12

Membrane Technology in Biological Wastewater Treatment

12.1 Introduction

The high performance of membrane technology has been proven in recent years in a wide range of fields, such as chemical industry, medical technology, drinking water treatment, biotechnology and environmental technology. The German work groups of the DWA (ATV-DVWK 2000a, b, 2002a, b; DWA 2005) have provided excellent reports on the application of membranes in the field of wastewater treatment. Also, the Chemical Engineering School at the RWTH Aachen has provided important knowledge of fundamentals and applications in the whole area of membrane technology (Rautenbach and Albrecht 1981, 1989; Melin and Rautenbach 2004).

The continuous development of membrane materials and membrane design on the one hand and the knowledge of operational management on the other hand have fostered the growth of membrane technology in wastewater treatment. Many questions have yet to be answered, however, especially in activated sludge systems:

- How can we best implement membranes in activated sludge systems?
- Do we need primary settlers if we use a membrane activated sludge process?
- Which membrane modules and operating modes are effective and energy efficient?
- Is it possible to perform nitrification and phosphate elimination in combination with membrane processes?

The number of membrane processes installed for the treatment of municipal wastewater is rather low but steadily increasing. In Germany, several large membrane processes for wastewater treatment plants (>11 000 inhabitants) are in operation or being planned (MUNLV 2003). The largest one (for 80 000 inhabitants; about 1900 m³ h $^{-1}$) has been in operation since 2004 in the wastewater treatment plant (WWTP) at Nordkanal (DWA 2005). The biggest industrial membrane process in biological wastewater treatment for a flow rate of about 200 m³ h $^{-1}$ has been in operation near Dortmund since 2004 for the treatment of wastewater from the pharmaceutical production plant of Schering AG (Achtabowski and Neuhaus 2005).

Fundamentals of Biological Wastewater Treatment. Udo Wiesmann, In Su Choi, Eva-Maria Dombrowski Copyright © 2007 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim ISBN: 978-3-527-31219-1

Industrial wastewater treatment plants are often characterized by low flow rates and high pollutant concentrations. The benefit of using membranes is the possibility to reuse water in different qualities and the gain of reusable material as well as environmental aspects integrated in the production process. Examples for treatment of water and wastewater with membranes are given in Table 12.1.

Membrane processes have different targets, such as meeting the guidelines of wastewater quality standards, the recovery of components or the reuse of the treated water. In the field of municipal wastewater treatment, micro- and ultrafiltration processes are predominantly applied with the target of avoiding the need for a secondary clarifier, increasing the bacterial concentration in activated sludge process or producing an effluent free from suspended solids. In industrial wastewater treatment and water reuse, nanofiltration and reverse osmosis are preferentially used.

Table 12.1 Examples of membrane processes in wastewater and polluted river water treatment (MUNLV 2003).

	Municipal, commercial and industrial applications
Micro- and ultrafiltration	MBR for treatment of: - municipal, pharmaceutical - rendering plant - food industry wastewater - landfill leachate Tertiary filtration of treated wastewater Treatment of distillery wastewater Treatment of electro dip coating water, oil-in-water emulsions and degreasing-processes in metal industries Reuse of concentrated water-soluble lacquers Pretreatment for production of boiler feed water Treatment of polluted river water with integrated precipitation Elimination of phosphate, iron and manganese
Nanofiltration	Elimination of specific compounds like EDS in wastewater Desalination of wastewater Discoloring of wastewater in textile and cellulose industries Concentration of landfill leachate
Reverse osmosis	Concentration of CaSO ₄ from mine-drainage water Recycling of silver from washing water in photo industry Treatment of wastewater in textile-dye industry Concentration of washing water in cellulose industries Reuse of phosphor acid Treatment of chlorine water Treatment of landfill leachate Desalination Reuse of water

12.2 Mass Transport Mechanism

12.2.1

Membrane Characteristics and Definitions

Membranes are flat, semi-permeable structures that are permeable for at least one component and are impermeable for others. According to the nomenclature of membrane technology, various membrane processes are characterized according to the molar mass or diameter of the transported component, the aggregate state on the two sides of the membrane as well as the separation principle. The transport can be caused by gradients of concentration or pressure. Pressure-driven membrane processes like micro-, ultra- and nanofiltration as well as reverse osmosis are used in wastewater treatment.

Figure 12.1 shows the classification of membrane processes based on the average particle diameter or molar mass, with a few examples of wastewater components. Note that the ranges of the separation processes overlap with respect to the particle diameter and the driving pressure.

The functional principle behind membrane processes used in wastewater treatment are filtration or sorption and diffusion, whereby the wastewater feed is divided into a cleaned part, i.e. the filtrate or permeate, and a concentrated part, i.e. the concentrate or retentate (Fig. 12.2).

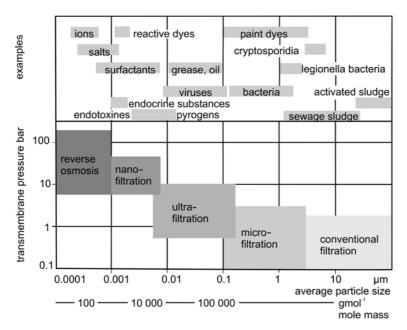


Fig. 12.1 Classification of membrane processes used in wastewater treatment (MUNLV 2003; Rautenbach 1997).

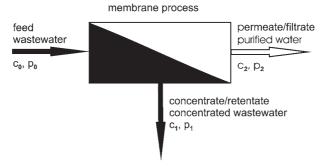


Fig. 12.2 Scheme showing the principle of the membrane process.

The performance of a membrane filtration unit is determined by the following main parameters:

• The selectivity of the membrane is the capability to separate between components like oil and water or salt and water. Low selectivity can only be compensated with an expensive multi-stage process. For aqueous systems of a solvent and a solute the retention coefficient or retention R is a measure of the selectivity. The solute is retained while the solvent, most often water, passes through the membrane; the retention R is given by:

$$R = \frac{c_0 - c_2}{c_0} = 1 - \frac{c_2}{c_0} \tag{12.1}$$

where c_0 is the concentration of the pollutant in the feed and c_2 is the concentration of the pollutant in the permeate.

The true retention achieved with the membrane is higher because the concentration of the retained component increases at the surface of membrane c_3 as a result of concentration polarization (Section 12.3).

$$R_{t} = 1 - \frac{c_2}{c_3} \tag{12.2}$$

In the field of biological wastewater treatment, one main component often has to be eliminated; and the feed and permeate concentrations are given, for example, as suspended solids in g $\rm L^{-1}$ MLSS.

• The relative volume flux J_{p0} characterizes the hydrodynamic permeability:

$$J_{p0} = \frac{Q_p}{\Delta p_{TM} A_m} m^3 m^{-2} h^{-1} bar^{-1}$$
 (12.3)

where Q_p is the permeate volume flow rate, Δp_{TM} is the transmembrane pressure and $A_{\rm m}$ is the membrane area.

• The gradient of trans-membrane pressure, i.e. the driving force, is given by:

$$\Delta p' = \frac{p_0 + p_1}{2} - p_2 \tag{12.4}$$

which takes into account the pressure drop along the cross-section of the membrane $p_0 - p_1$.

• Mechanical stability and resistance to fouling and scaling must be considered as other important factors.

Low permeability of a given membrane can be compensated by increasing the membrane surface area. The permeate flux J_p or the permeate velocity w_p is given by:

$$J_{p} = w_{p} = \frac{Q_{p}}{A_{m}} m^{3} m^{-2} h^{-1}$$
 (12.5)

The flux and the retention coefficients R and Rt are not constant along the surface area of a membrane, even if there is no variability in the quality of the membrane material. The concentration of the retained component increases continuously and affects the flux and retention coefficients.

In wastewater treatment, transmembrane pressure Δp_{TM} varies from 0.1 bar up to 120 bar. The characteristic cut-off of a membrane corresponds either to the particle diameter (in microns) or to the molar mass (measured in Dalton) of the largest retained substance.

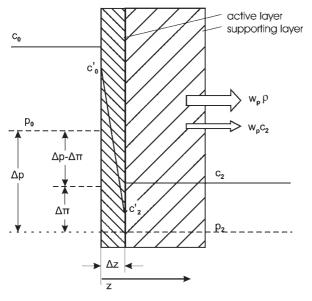


Fig. 12.3 Concentration and pressure gradients through a solution-diffusion membrane. We have to distinguish $\Delta p'$ of Eq. (12.4) from Δp of Figure 12.3.

The cut-off of a membrane is determined as the molar mass of macro-molecules and dissolved substances with a retention coefficient of 90% or 95%. It is determined experimentally by the fractional separation curves for ultrafiltration membranes with different substances (Rautenbach 1997) and is often used for the characterization of membrane processes with the exception of microfiltration.

Various transport models are employed when studying the selectivity of different membranes and their transport mechanisms (Rautenbach 1997):

- A black-box model resulting from a large base of experimental results from real systems of combinations of treated fluids and membranes.
- Semi-empirical models for the real system with regard to physical and chemical parameters (solution-diffusion and pore model).
- Structural models in fundamental research.

Here we use semi-empirical models because it is common engineering practice to utilize the understanding of physical properties together with results of investigating process parameters. The solution-diffusion model (reverse osmosis and partly nanofiltration) and the pore model (ultra- and microfiltration) can be used with the physical and chemical background information to reduce the required number of experiments for studies and to quantitatively optimize membrane filtration operation in the area of wastewater treatment.

The design and layout of biological wastewater treatment plants with membrane bioreactor (MBR) technology has to focus on the requirements of the activated sludge process. The first guidelines were formulated by ATV-DVWK (2000b) with special regard to reactor volume, oxygen transfer rate, pre-treatment of wastewater, sludge disposal and, of course, membrane performance and cleaning (Section 12.3).

First we will focus on the most common mechanistic models of mass transport through membranes. They are based on diffusion and convection. Then we will consider resistances to mass transfer, like concentration polarization as well as the combination of transport mechanisms and resistances. A further combination of models, e.g. the solution-diffusion model and the pore model, is necessary when using membranes with an active, deep layer, a porous carrier layer or if a gel-layer is formed.

12.2.2

Mass Transport Through Non-porous Membranes

The solution-diffusion model describes reverse osmosis in ideal membranes as well as nanofiltration processes in non-porous membranes.

The transport of a component through a non-porous membrane is only possible by dissolution and diffusion. In this idealized model of mass transport, the membrane is treated like a liquid. Figure 12.3 shows the concentration and pressure gradients for an asymmetric membrane, where the active layer is responsible for separation and the supporting layer for mechanical stability.

The specific mass transfer rate of diffusion is described by Fick's first law:

$$J_{\rm D} = -D\nabla c' \tag{12.6}$$

where c' is the concentration of the dissolved component and D is the diffusion coefficient.

The diffusion coefficient D is idealized to be independent of concentration. That means D is constant and not affected by location in the membrane or by concentration. For one-dimensional diffusion, the diffusive flux follows as:

$$J_{\rm D} = -D \frac{dc'}{dz} g m^{-2} h^{-1}$$
 (12.7)

or (see Fig. 12.3):

$$J_{\rm D} = -D \frac{c_0' - c_2'}{\Lambda_{\rm Z}} \tag{12.8}$$

This is a solution for the steady-state mass balance for diffusion across two planes (e.g. through a thin plate membrane at the points z and z + dz):

$$0 = -\frac{\mathrm{d}J_{\mathrm{D}}}{\mathrm{d}z} \tag{12.9}$$

and considering Eq. (12.7):

$$0 = \frac{d}{dz} \left(D \frac{dc'}{dz} \right) = D \frac{d^2c'}{dz^2}$$
 (12.10)

According to Henry's law, we obtain for ideal systems with linear sorption characteristics with respect to the dissolved impurity concentration in the membrane c':

$$c = Hc' (5.6)$$

$$J_{D} = \frac{D}{\Delta z H} (c_{0} - c_{2}) = B (c_{0} - c_{2})$$
(12.11)

with:

$$B = \frac{D}{\Delta z H} \tag{12.12}$$

The membrane constant for the dissolved impurity B is independent of pressure but is a function of temperature because H and D vary with temperature.

The diffusive water flux follows in analogy to (12.8):

$$J_{\rm DW} = D_{\rm W} \, \frac{c'_{\rm 0W} - c'_{\rm 2W}}{\Delta z} \tag{12.13}$$

Because of the water surplus, the absorption equilibrium can be described via (partial) pressure:

$$c'_{W} = H'_{W} p$$
 (12.14)

and the water flux follows taken together with Eq. (12.13):

$$J_{\rm DW} = \frac{D_{\rm W} H_{\rm W}'}{\Lambda_{\rm Z}} (p_{\rm o} - p_{\rm 2}) \tag{12.15}$$

Usually, the water flux is written as a function of the pressure difference:

$$J_{DW} = \rho A (p_0 - p_2) \tag{12.16}$$

with:

$$A = \frac{D_{\rm w} H_{\rm w}'}{\rho \Delta z} \tag{12.17}$$

as the membrane constant A and ρ as the density of water. Note that, in contrast to the dimensionless H (see Eq. 5.6), H'_{w} has the dimension g (m³ bar)⁻¹ (Eq. 12.14).

The membrane constant A for the solvent (here water) is a function of temperature as well as a function of membrane properties $[D_w(T), H'_w(T)]$ and the thickness of the active layer Δz .

Equation (12.16) is valid if the osmotic pressure Π is negligible. Otherwise the diffusive flux is:

$$J_{\rm DW} = \rho \, A \, (p_0 - p_2 - \Delta \Pi) \tag{12.18}$$

with:

$$\Pi = -\frac{RT \ln a_i}{V_i} \tag{12.19}$$

it follows:

$$\Delta\Pi = \frac{RT}{V_i} (\ln a_{i,o} - \ln a_{i,2})$$
 (12.20)

$$a_i = \gamma_i x_i = \gamma_i \left(1 - x_i \right) \tag{12.21}$$

where a_i is the solvent activity, γ_i is the activity coefficient, x_i is the mole fraction, x_i is the mole fraction of the solute, V_i is the molar volume of the solvent and R is the gas constant.

For ideal mixtures or for very low solute concentrations (x_i<<1) the activity coefficient y_i is unity (Mulder 2000) and the activity becomes:

$$\ln a_i = \ln x_i = \ln (1 - x_i) \approx -x_i$$
 (12.22)

Thus, using the equation for the state of ideal gases for diluted solutions the molar concentration follows according to the amount of moles N_i and N_{Σ} :

$$c_{j} = \frac{N_{j}}{V} = \frac{x_{j} N_{\Sigma}}{V} \approx \frac{x_{j}}{V_{j}}$$
(12.23)

At least the osmotic pressure difference between membranes follows:

$$\Delta\Pi = RT(c_0 - c_2) \tag{12.24a}$$

respectively for dissociating compounds, including the osmotic coefficient β:

$$\Delta\Pi = \beta RT (c_0 - c_2) \tag{12.24b}$$

where c_0 is the feed molar concentration of the solute, c_2 is the permeate molar concentration of the solute and β is the osmotic coefficient for the change in numbers of moles by dissociation, depending on the degree and stoichiometrics of the dissociation reaction (Rautenbach and Albrecht 1981).

From Eqs. (12.23) and (12.24) it follows that $\Delta\Pi$ is high for high values of c_0 and for low values of c2. Considering concentration polarization, the solute concentration at the membrane surface is $c_3 > c_0$ (Section 12.3).

The membrane constants A and B depend on temperature according to exponential functions (Rautenbach 1997) because of its influence on D and H (B in Eq. 12.12) and on D_w and H'_w [A in Eq. (12.17)]. Yet the temperature dependency of B is often neglected for practical purpose. A and B have to be determined experimentally. They characterize the permeability of the membrane; the quotient A/B is a degree for the selectivity of the system. For a low A/B the selectivity is high.

Often the designer's aim is to calculate:

- The flow rate of the water through membrane w_p.
- The permeate concentration c₂.

With the flow rate of permeate W_p (Fig. 12.3), we obtain the mass flux J_D for the solute with c2:

$$J_{\rm D} = W_{\rm p} c_2 \tag{12.25}$$

and for water:

$$J_{\rm DW} = W_{\rm p} \rho \tag{12.26}$$

From Eqs. (12.18) and (12.26), the permeate flow rate can be calculated:

$$W_{p} = \frac{J_{DW}}{\rho} = A (p_{0} - p_{2} - \Delta \Pi)$$
 (12.27)

From Eqs. (12.25) and (12.11), considering the flow rate according to Eq. (12.27), we obtain:

$$c_{2} = \frac{J_{D}}{w_{p}} = \frac{B(c_{0} - c_{2})}{A(p_{0} - p_{2} - \Delta\Pi)}$$
(12.28)

and finally the result for the permeate concentration c2:

$$c_2 = \frac{B c_0}{A (p_0 - p_2 - \Delta \Pi) + B}$$
 (12.29)

For $p_0 - p_2 = \Delta \Pi$, no water is transported through the reverse osmosis membrane but the dissolved compound is transported, giving finally $c_2 = c_0$.

At the membrane surface, some important effects may occur, e.g. concentration polarization and fouling processes that have to be taken into consideration (Section 12.3). In reverse osmosis and nanofiltration processes osmotic pressure is important, but even in ultrafiltration processes osmotic pressure can have an effect. At high fluxes and high retention values, the concentration of macromolecules at membrane surface becomes quite high (see Problem 12.1).

Mass Transport Through Porous Membranes

The pore model is based on the assumption that the membrane pores are much smaller than the diameter of the retained particles but are permeable to water. In contrast to the solution-diffusion model, water does not diffuse through the pores; rather it flows under the influence of pressure and frictional forces. The pore system is ideally straight and parallel; and each pore has the same circular size. As a result of the small capillary diameter, laminar flow conditions are given and Hagen-Poiseuille's law is valid. For pores with diameter d and length L, one can write in dimensionless notation:

$$\xi = \frac{64}{Re} \tag{12.30}$$

with Reynolds number:

$$Re = \frac{w_{po} d}{v} \tag{12.31}$$

and resistance number ξ :

$$\xi = \frac{\Delta p}{\frac{1}{2} \rho w_{po}^2} \frac{d}{L}$$
 (12.32)

with w_{po} as the flow rate in a capillary pore.

In reality, the length of pores L is greater than the membrane thickness Δz . Therefore, a mean tortousity μ is defined:

$$\mu = \frac{L}{\Lambda_7} \tag{12.33}$$

The flow rate of permeate w_p is now given by Hagen-Poiseuille's law (using Eqs. 12.30 to 12.33) with porosity ε :

$$w_{p} = w_{po} \epsilon = \frac{\Delta p}{32 \, n} \frac{d^{2} \epsilon}{\mu \Delta z} \tag{12.34}$$

Carman and Kozeny's pore model (Carman 1956) assumes a pore system formed of equally sized spheres in a packed bed. With permeate flux:

$$J_{p} = w_{p} = \frac{\Delta p \,\varepsilon^{3}}{2 \,\eta \,(1 - \varepsilon)^{2} \,a_{v}^{2} \,\mu \Delta z} \tag{12.35}$$

$$a_{v} = \frac{A_{po}}{V} \tag{12.36}$$

the hydraulic diameter d_h for packed beds is introduced (Rautenbach and Albrecht

$$d_{h} = \frac{4\varepsilon}{(1-\varepsilon) a_{v}} \tag{12.37}$$

where a_v is the volume-specific surface area.

Equation (12.35) is known as the Carman-Kozeny equation. For sphere-packed beds, the tortousity is given as μ =25/12 (Rautenbach and Albrecht 1989). With the membrane constant A*:

$$A^* = \frac{\varepsilon^3}{2\eta (1-\varepsilon)^2 a_v^2 \mu \Delta z}$$
 (12.38)

the equation shows the linear dependence of permeate flux on the driving force, i.e. the pressure gradient across the membrane:

$$J_{p} = A^{*} \Delta p \tag{12.39}$$

Comparing the diffusive water flux according to Eq. (12.16), A* corresponds to the product of the density and the membrane constant A used in the solution-diffusion model.

The linear behavior of J_p in relation to Δp depends only on the membrane constant A* of a given membrane. A* must be determined experimentally because it depends on viscosity, which is a function of temperature. Figure 12.4 shows the membrane-controlled flux.

This model fits micro- and ultrafiltration processes, but in practice a gel layer at the surface of the membrane and a depositing of particles also affects mass transport (see Section 12.3).

12.3 Mass Transfer Resistance Mechanisms

12.3.1

Preface

In pressure-controlled membrane filtration (e.g. in the field of wastewater treatment), mass transfer across the membrane is affected by multiple resistances before and behind the membrane surface. In the case of asymmetric membranes used in ultrafiltration and reverse osmosis, one has a combination of local transport resistances. The resistance of the active layer is mostly rate-limiting. In reverse osmosis and in some cases of ultrafiltration, permeate fluxes are low and concentration polarization is insignificant because of the high diffusive back-flow of the small molecules. All components, even those at the surface of the membrane, remain soluble; here the membrane itself controls the mass transfer.

In every case where a concentration polarization profile can occur the effect can be controlled more or less by the flow conditions along the surface of membrane and it is therefore influenced by membrane module design (Section 12.4).

We will now discuss resistances to mass transfer and the most common model for the calculation of the concentration c₃ at the membrane surface in combination with mass transport mechanisms for solution-diffusion and the pore model.

12.3.2

Mass Transfer Resistances

Membrane-controlled mass transfer provides the simplest operational condition for porous membrane systems with pressure as the driving force. Equation (12.39) is often written in the form of Darcy's Law (Darcy 1856):

$$J_{p} = \frac{\Delta p}{\eta R_{m}} \tag{12.40}$$

with:

$$R_{\rm m} = \frac{2(1-\epsilon)^2 a_{\rm v}^2 \mu \Delta z}{\epsilon^3} = \eta \cdot A^{*-1} m^{-1}$$
 (12.41)

for laminar flow as resistance of membrane $R_{\rm m}$. $J_{\rm p}$ is only controlled by the clean, non-blocked membrane and shows linear behavior (Fig 12.4). A further hydraulic resistance due to pore blocking and adsorption $R_{\rm f}$ can be considered:

$$J_{\rm p} = \frac{\Delta p}{\eta \left(R_{\rm m} + R_{\rm f} \right)} \tag{12.42}$$

During a membrane filtration process, the solute concentration at the membrane surface rises; and when the concentration c_3 exceeds the solubility limit crystallization occurs. A gel layer, also known as a sludge cake, is formed by the deposit of solids as well as by the growth of bacteria (see Fig. 12.5 in the next section and Fig. 12.9 in Section 12.4.3). Practice normally shows non-linear behavior because the gel layer controls mass transport (Fig. 12.4).

Gel layer or sludge cake formation is often found above a so-called critical flux (see Section 12.5). The accumulated material simply adds a further resistance $R_{\rm c}$ for the sludge cake to the resistance of the free or blocked membrane:

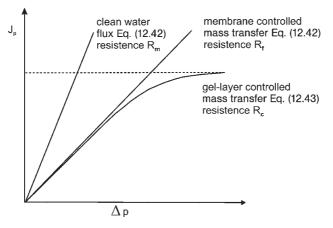


Fig. 12.4 Permeate flux plot as a function of pressure difference.

$$J_{p} = \frac{\Delta p}{\eta (R_{m} + R_{f} + R_{c})}$$
 (12.43)

respectively:

$$J_{p} = \frac{\Delta p}{\eta R_{\Sigma}} \tag{12.44}$$

12.3.3

Concentration Polarization Model

The classic concentration polarization model describes the fluxes according to convective flow and retransfer of the solute, i.e. back diffusion through concentration boundary layer dependent on the concentration gradient between the bulk region and the surface of the membrane (Fig. 12.5).

One solution to the mass balance in the boundary layer with thickness δ is obtained at steady state conditions, $dJ_{\Sigma}/dz = 0$. There are two parts of the total flux: the flux of the solute counter to the z-direction, $-w_{\scriptscriptstyle p} c,$ and the back diffusion of the solute, -D dc/dz. Assuming that the permeate velocity is independent of the z-coordinate it follows that:

$$J_{\Sigma} = -w_{p}c - D\frac{dc}{dz}$$
(12.45)

After differentiation of Eq. (12.45), we obtain at steady-state conditions:

$$0 = w_p \frac{dc}{dz} + D \frac{d^2c}{dz^2}$$
 (12.46)

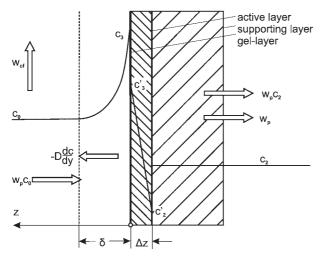


Fig. 12.5 Concentration polarization on the feed side and boundary layer.

A possible solution of Eq. (12.46) is the exponential function:

$$c = a_1 + a_2 \exp(a_3 z) \tag{12.47}$$

and after differentiating Eq. (12.47):

$$\frac{\mathrm{dc}}{\mathrm{dz}} = a_3 a_2 \exp(a_3 z) \tag{12.48}$$

$$\frac{d^2c}{dz^2} = a_3^2 a_2 \exp(a_3 z) \tag{12.49}$$

Three boundary conditions are necessary to determine the constants a₁, a₂ and a₃. According to Fig. 12.5: (1) at the boundary layer with thickness δ , the concentration is the same as in the bulk flow c_0 , (2) the highest concentration c_3 is attained at the surface of membrane at z = 0, and finally, (3) the condition for the permeate flux of the solute is the equality of the sum of convective flux and the back diffusive flux at the membrane surface:

$$z = \delta, \quad c = c_0 \tag{12.50}$$

$$z = 0, \quad c = c_3$$
 (12.51)

$$z = 0, -w_p c_2 = -w_p c_3 - D \left. \frac{dc}{dz} \right|_{z=0}$$
 (12.52)

From Eqs. (12.48) and (12.49) in Eq. (12.46) we obtain:

$$0 = w_p a_3 a_2 \exp(a_3 z) + D a_3^2 a_2 \exp(a_3 z)$$
 (12.53)

and finally for the constant a3:

$$a_3 = -\frac{w_p}{D} {(12.54)}$$

From Eqs. (12.52) and (12.51) we obtain:

$$-w_{p}c_{2} = -w_{p}c_{3} - D a_{3}a_{2}$$
 (12.55)

or with Eq. (12.54):

$$a_2 = c_3 - c_2 \tag{12.56}$$

Finally, considering condition Eq. (12.50), then Eq. (12.47) gives:

$$c_0 = a_1 + (c_3 - c_2) \exp\left(-\frac{w_p}{D}\delta\right)$$
 (12.57)

respectively:

$$a_1 = c_0 - (c_3 - c_2) \exp\left(-\frac{W_p}{D}\delta\right)$$
 (12.58)

Now, all three integration constants are known and the concentration profile near membrane $(0 \le z \le \delta)$ follows to:

$$c = c_0 - (c_3 - c_2) \exp\left(-\frac{w_p}{D}\delta\right) + (c_3 - c_2) \exp\left(-\frac{w_p}{D}z\right)$$
 (12.59)

Considering Eq. (12.51), a simple result describes the dimensionless concentration polarization as a function of velocity w_p and diffusion coefficient D of the retained component at the membrane surface z = 0 according to Eq. (12.59):

$$\frac{c_3 - c_2}{c_0 - c_2} = \exp\left(\frac{w_p}{D}\delta\right) \tag{12.60}$$

The permeate flow rate w_p is then given by:

$$w_{p} = k_{L} \ln \left(\frac{c_{3} - c_{2}}{c_{0} - c_{2}} \right)$$
 (12.61)

with the mass transfer coefficient:

$$k_{L} = \frac{D}{\delta} \tag{12.62}$$

The mass transfer coefficient can be expressed by dimensionless numbers with $w_{\rm cf}$ as the cross-flow rate:

Sherwood number:
$$Sh = \frac{k_L d}{D}$$
 (12.63)

Schmidt number:
$$Sc = \frac{V}{D}$$
 (12.64)

Reynolds number:
$$Re = \frac{w_{cf} d}{v}$$
 (12.65)

These numbers and L/d result from a dimensional analysis of the problem. The determination of the relation between the Sherwood, Schmidt and Reynolds numbers as well as L/d is based on the analogy between heat and mass transport (Table 12.2). For laminar flow and simple geometric conditions, they can be calculated by solving mass and momentum balances. The relations presented are

Table 12.2 Sherwood number for different flow conditions experimentally proved for mass transport by Linton and Sherwood (1950) and Rautenbach and Albrecht (1981).

	Sherwood number	Flow condition
Tube	$Sh = 1.62 (Re Sc d/L)^{1/3}$ $Sh = 0.023 Re^{7/8} Sc^{1/4}$ $Sh = 0.04 Re^{3/4} Sc^{1/3}$	Laminar Turbulent Turbulent
Canal with rectangular cross-section	$Sh = 1.85 \; (Re \; Sc \; d_{\rm h}/L)^{1/3} \label{eq:Sh}$ $Sh = Sh_{\rm tube} \label{eq:Sh}$	$\begin{array}{l} Laminar \\ Turbulent \\ Re \ Sc \ d_{\rm h}/L > 10^2 \end{array}$

proven experimentally for mass transport by convection and diffusion in tubes for laminar and turbulent flow conditions (Linton and Sherwood 1950) and in channels for laminar flow conditions (Rautenbach and Albrecht 1981).

From steady-state laminar flow in tubes of diameter d and length L as well as d/L≪1:

Sh = 1.62
$$\left(\text{Re Sc } \frac{d}{L} \right)^{1/3}$$
 (12.66)

is valid. After calculation of k_L from Eq. (12.66) and introduction into Eq. (12.61), the permeate rate follows:

$$w_{p} = 1.62 \left(\frac{D^{2} w_{cf}}{dL} \right)^{1/3} \ln \left(\frac{c_{3} - c_{2}}{c_{0} - c_{2}} \right)$$
 (12.67)

We must determine not only the concentrations but also the diffusive coefficient D. A possible approximation for D is given by the Stokes Einstein equation in Eq. (5.4) as a function of absolute temperature T, solute radius R, dynamic viscosity η and with the Boltzmann constant K (Section 5.1; Mulder 2000):

$$D = \frac{KT}{6\pi nR}$$
 (5.4)

Deviations arise if D and v become dependant on concentration, as observed in the ultrafiltration of macromolecules, or when the cross flow rate rises for small values of d/L (Rautenbach 1997). This occurs particularly in ultrafiltration processes in the field of wastewater treatment if suspended solids are present which accumulate at the boundary layer.

12.3.4

Solution-diffusion Model and Concentration Polarization

The aim is now to calculate w_p and c_2 as well as $\Delta\Pi$. Thus, we need three independent equations. Considering the increasing concentration c3 at the membrane surface for the calculation of $\Delta\Pi$ and J_D , we obtain Eqs. (12.68) and (12.69) from Eq. (12.24b), respectively Eq. (12.11) and (12.12).

$$\Delta\Pi = \beta RT (c_3 - c_2) \tag{12.68}$$

$$J_{D} = B(c_{3} - c_{2}) \tag{12.69}$$

For the solution diffusion model (reverse osmosis and nanofiltration) w_p follows according to Eqs. (12.27) and (12.68):

$$W_{p} = A (p_{0} - p_{2} - \beta RT (c_{3} - c_{2}))$$
(12.70)

Equations for the concentration of the retained component c₃ and the permeate c₂ are given for c_2 according to Eqs. (12.29) and (12.27):

$$c_2 = \frac{B c_3}{w_p + B} \tag{12.71}$$

The third independent equation is given by elimination of c_3 from Eq. (12.61):

$$c_3 = c_2 + (c_0 - c_2) \exp \frac{W_p}{k_1}$$
 (12.72)

After introducing Eq. (12.72) in Eq. (12.71), c_3 and c_2 can be eliminated:

$$c_{2} = \frac{B c_{0} \exp \frac{W_{p}}{k_{L}}}{w_{p} + B \exp \frac{W_{p}}{k_{L}}} = \frac{B c_{0}}{w_{p} \exp \left(-\frac{W_{p}}{k_{L}}\right) + B}$$
(12.73)

 c_3 follows from Eq. (12.72).

With Eqs. (12.72) and (12.73) in Eq. (12.70), we finally obtain an implicit equation

$$w_{p} = A \left(\Delta p - \beta RT \frac{w_{p} c_{0} \exp \frac{w_{p}}{k_{L}}}{w_{p} + B \exp \frac{w_{p}}{k_{L}}} \right)$$

$$(12.74)$$

Equation (12.74) can only be solved by graphical or numerical methods after determination of k_L using one of the empirical equations in Table 12.3 (see Section 12.4.1). With these solutions, c_3 and c_2 can be calculated using Eqs. (12.72) and (12.73).

There is, however, a correlation between the true retention coefficient R_t and c₃, the concentration at the surface of the membrane according to Eq. (12.2). With Eqs. (12.70) and (12.71), we obtain the true retention coefficient R_t which can be calculated using Eqs. (12.72) and (12.73):

$$R_{t} = 1 - \frac{B}{A(p_{0} - p_{2} - \beta RT(c_{3} - c_{2})) + B}$$
(12.75)

Explicit solutions of Eq. (12.74) are possible for some specific cases:

1. If concentration polarization does not occur, $c_3 = c_0$, $\exp(w_p/k_L) = 1$ follows from Eqs. (12.60) and (12.62). Solving the quadratic equation we obtain:

$$w_{p} = A \left(\Delta p - \beta RT \frac{w_{p} c_{0}}{w_{p} + B} \right)$$
 (12.76)

2. Furthermore, if the membrane is not permeable for the solute (B = 0), the permeate specific flow rate is:

$$w_p = A \left(\Delta p - \beta RT c_0 \right) \tag{12.77}$$

3. In the case of low feed concentration c_0 , then $\Delta\Pi$ can be neglected:

$$W_{p} = A \Delta p \tag{12.78}$$

12.3.5

The Pore Model and Concentration Polarization

The permeate flow rate is determined with the pore model via the resistance in series models, as described in Sections 12.3.2.

Different authors use the resistance in a series of models and describe mathematical approaches as well as experimental investigations for estimation of the resistances used in Eq. (12.43) (Fu and Dempsey 1998; Melin et al. 2001; Geissler et al. 2003; Wintgens et al. 2003). For example, resistances were summarized by Chang et al. (2002) for microfiltration in MBR with $R_{\rm m} = 3 - 22 \cdot 10^{11} \, {\rm m}^{-1}$, $R_{\rm c} = 3 - 48 \cdot 10^{11} \, {\rm m}^{-1}$ and $R_{\rm f} = 10^{-11} \ m^{-1}$ for different membrane materials. In ultrafiltration processes for the reuse of surfactants $R_{\rm m}$ =5 \cdot 10¹² m⁻¹ and $R_{\rm f}$ =5-9.1 \cdot 10¹² m⁻¹ was experimentally determined for PES membranes for various surfactants (Goers 2000).

The resistance of the sludge cake R_c is a function of the concentration at the membrane surface. It follows from Eq. (12.43) according to the concentration polarization model.

Chang and Fane (2000) describe the shear stress on micro and ultrafiltration membrane surfaces from air bubbling of submerged hollow fibers by slug flow conditions, i.e. based on two-phase flow conditions influenced by the formation of Taylor bubbles. Melin et al. (2001) and Wintgens et al. (2003) use the same model of slug flow conditions, considering concentration polarization and hydrodynamic effects. They use experimental data from the full-scale wastewater treatment plant Rödingen in Germany to obtain coefficients for their model. Further investigations are summarized by Stephenson et al. (2000).

Performance and Module Design

12.4.1

Membrane Materials

The efficiency of membrane filtration processes in wastewater treatment is decisively influenced by the selection of the membrane material for the wastewater to be treated with regard to the particles and dissolved compounds it contains. Therefore, the membrane material has to be chosen carefully by the supplier in cooperation with the plant engineers. It is usually necessary to perform investigations in laboratory or pilot scale and to test different membrane materials and modules. Table 12.3 presents an overview of common membrane materials.

Depending on the composition and properties of the wastewater to be treated as well as on the membrane's mechanical stability, it is possible to use either organic or inorganic solid materials. Once commonly used, cellulose membranes are now used less often in recent times compared to polymer membranes, like polysulfone PS, polyacrylonitrile PAN and polyethersulfone PES. Because of their resistance to high temperatures and chemical stress, the use of inorganic materials like ceramics, aluminum, refined steel and glass is becoming more important (MUNLV 2003).

Polyamide PA 95% Cellulose acetate AC 5%

	Membrane structure	Membrane material	Active layer material
Microfiltration	Symmetric, porous	Polymer and ceramic	Polypropylene PP Polyvinylidenfluoride PVDF Polysulphone PSU Aluminium oxide Refined steel Titanium dioxide Zirconium dioxide
Ultrafiltration	Asymmetric, porous	Polymer phase inversion, composite membrane and ceramic	Polysulphone PSU Reg. cellulose Polyacrylnitrile PAN Polyethersulphone PES Titanium dioxide Zirconium dioxide Polyvinylidenfluoride PVDF
Nanofiltration	Asymmetric, dense	Polymer phase inversion, composite membrane	Polyamide PA (Zirconium dioxide) Polyethersulphone PES Cellulose acetate CA

Table 12.3 Common membrane materials and membrane structures used in membrane processes (according to ATV-DVWK 2002a).

Membranes are either symmetric or asymmetric in structure. That means they exhibit a homogenous or inhomogeneous dispersion of material. An asymmetric membrane is constructed by two layers of one material with different porosities (a phase inversion membrane), or by two layers of different materials (a composite membrane).

Polymer phase inversion,

composite membrane

The very thin and dense active layer of about 1 μm on the feed side is responsible for the membrane's performance and permeability. The porous second layer is the supporting structure. Higher fluxes are attainable, especially for solution diffusion membrane processes, with asymmetric membranes. Both composite and phase inversion membranes can be constructed as porous or non-porous membranes (Rautenbach 1997).

12.4.2 Design and Configuration of Membrane Modules

Asymmetric,

dense

12.4.2.1 Preliminary Remarks

Reverse osmosis

The following informations are valid generally for all possible applications. There are several very different ways to construct membrane modules by the arrangement of the inlet and outlet streams. For example, a three-end module for cross-flow configuration consists of a frame construction for the selected membrane

Table 12.4 Characteristics, advantages and disadvantages of plate and tubular membrane modules (MUNLY 2003; VDMA 2005).

	Tubular membranes			Plate membranes		
	Tubular module	Capillary module	Hollow fiber module	Plate and frame module	Spiral wound module	Membrane cushion
Active layer	Inside	Out-/inside	Out-/inside	Outside	Outside	Outside
Internal diameter Membrane surface	5.5–25 mm ca. $80 \text{ m}^2 \text{ m}^{-3}$	$0.25-5.5 \text{ mm}$ ca. $1000 \text{ m}^2 \text{ m}^{-3}$	$0.04-0.25 \text{ mm}$ ca. $10000 \text{ m}^2 \text{ m}^{-3}$	$40-100 \text{ m}^2 \text{ m}^{-3}$	ca. $1000 \text{ m}^2 \text{ m}^{-3}$	$400 \mathrm{m^2 m^{-3}}$
area–volume ratio Configuration	Cross flow	Dead end/cross	Dead end	Cross flow	Dead end/cross	Dead end/cross
Advantages	Low blocking, low pressure drop, gel layer controlled	High surface area-volume ratio, low design costs, permeate back-	Very high surface area-volume ratio, low specific membrane costs,	Simple exchange of membranes, low blocking	High surface area-volume ratio, fewer seals, low design costs	Low pressure drop, low fouling
Disadvantages	Low surface area–volume ratio	washing Low compression strength	stable against high pressure Blockage, pressure drop	Many seals, low surface area-volume ratio	Long permeate ways, no mechanical cleaning, high blockage danger	Low surface area–volume ratio, many seals
					DOCKAGO CALLEST	

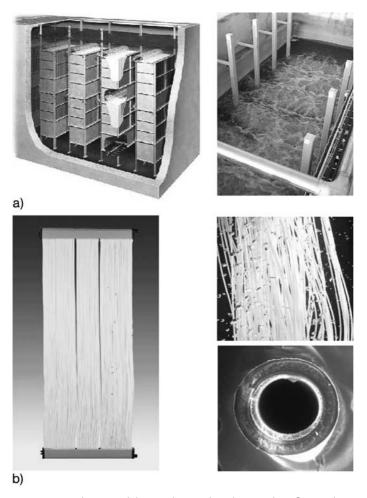


Fig. 12.6 Membrane modules in submerged mode. (a) Kubota flat panel, submerged in an unit with bubble diffusor. (b) Zenon hollow fiber module, with air bubbling and a hollow fiber in detail.

material with connections to one inlet (feed) and two outlet streams (permeate and concentrate) and a pump providing the driving pressure. Modules with tubular or plate membranes are described in Table 12.4.

Five principle configurations are commonly found (Stephenson et al. 2000):

- Plate-and-frame modules, commonly known from electrolysis stacks, are often used for micro- and ultrafiltration and less often for reverse osmosis (e.g. Kubota flat panel; Fig. 12.6a). Similarly, there are membrane cushions with permeate spacers and support plates welded together with the plate membranes.
- Spiral-wound modules are the standard configuration for reverse osmosis and nanofiltration modules.

- Membrane cushions are although used in ultra- and nanofiltration modules.
- Hollow fine fiber (Zenon; Fig. 12.6b) and capillary membranes are self-supporting membranes with out-to-in flow direction for hollow fibers and in-to-out flow direction for capillary ones.
- Tubular membranes are used for high turbulence and good cleaning characteristics (Berghof 2005; Fig. 12.7a). An example for a complete process is the Wehrle ultrafiltration plant in Fig. 12.7b.

Module design has to fulfil the following requirements:

- High ratio of membrane area to module bulk volume.
- Low pressure drop, low energy demand.
- High degree of turbulence on the feed side to promote mass transfer.
- Low costs per unit membrane area.
- Good cleaning management.





a)



b)

Fig. 12.7 Membrane modules in cross-flow mode. (a) Berghof tubular module and membranes. (b) Wehrle, MBR, ultrafiltration plant for the food industry.

For the treatment of a feed with a high suspended solids concentration, the demands for a high membrane area: volume ratio together with a design that facilitates cleaning is inconsistent and a compromise is necessary. Furthermore, hydrophilic behavior, attainable permeate volume flux, cut-off and operational life of the membrane have to be taken into account. Configurations such as dead-end, submerged and cross-flow mode are used for ultra- and microfiltration.

12.4.2.2 **Dead-end Configuration**

In the dead-end filtration process, the flow of the wastewater stream is orthogonal to the membrane and no retentate stream is produced. A two-end module is needed (Fig. 12.8a).

As a consequence, the retained colloids and macromolecules build a layer on the membrane surface which has to be removed periodically. This configuration results in an unsteady process which demands a two-end module and is only suitable if the feed suspended solids concentration is low.

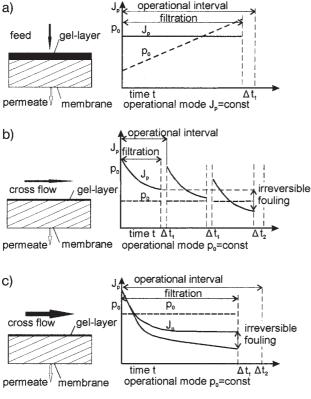


Fig. 12.8 Configuration of membrane filtration modules: (a) dead-end mode, (b) submerged mode, (c) cross-flow mode. $\Delta t_1 = \text{backwashing period}; \Delta t_2 = \text{chemical enhanced cleaning period}.$

12.4.2.3 Submerged Configuration

A special configuration of a two-end module is the submerged mode because a cross flow near membrane surface is applied without producing an additional stream (Fig. 12.8b). To minimize the building of a sludge cake layer and to avoid the high energy cost for high turbulence cross flow, a submerged system is used. The membranes are immersed directly in the activated sludge tank, where intensified aeration produces cross flow at the membrane surface (see Fig. 12.6b). Investigations have been performed on the influence of shear stress induced by bubbles (see Section 12.3.5). Permeate is removed by vacuum at transmembrane pressures of about 0.05 bar to 0.6 bar (Günder 1999; ATV-DVWK 2002b).

Transmembrane pressure is remarkably lower than 1 bar because of the drop in pressure which results from the water column above the submerged membrane. The rejected biomass remains in the bioreactor and the purified water passes through the membranes, usually from the outside in. Hollow fibers are flexible and move slightly to reduce gel layer formation because they are slightly longer than the module length and are therefore free to move (see Fig. 12.8b).

One significant characteristic of the submerged system is its operation in quasi steady state, in contrast to dead-end mode, and its higher frequency of back washing. Compared to the cross-flow configuration, there is less energy used (per m³ permeate) but a greater membrane surface area is needed because of the low attainable fluxes in submerged systems. Recently, submerged systems have become highly significant in the field of aerobic biological wastewater treatment. The advantages and disadvantages of dead-end and submerged mode are summarized in Table 12.5.

Enhanced permeate flux was proven to be a result of two-phase flow. Gassparged ultrafiltration with tubular membranes has been investigated experimentally with different suspensions as well as with river water (Chang and Fane 2000; Cabbasud et al. 2001) although computational fluid dynamics (CFD) were used to model this two-phase flow conditions (Taha and Cui 2002).

12.4.2.4 Cross-flow Configuration

In cross-flow filtration, the wastewater flow is parallel to the membrane (Fig. 12.8c). There is a continuous retentate stream in addition to the feed and permeate stream; and a three-end module is necessary. The retained material builds a layer on the surface of the membrane which can be influenced by the cross-flow rate. Thus the process is in continuous operation (see Table 12.5).

The reversible gel layer can be controlled by cross-flow conditions, while an irreversible gel layer has to be removed by a cleaning procedure. Gel layer-controlled filtration helps to protect microfiltration membranes from pore blockage. In general, there are two methods to control the severity of the reversible gel layer. When high turbulence at the membrane surface is realized by higher cross-flow rate with or without air injection, it leads to high energy costs. Higher turbulence can be obtained, though, by the use of smaller hydraulic diameters of membrane canals in tubular or flat membranes. This, however, increases the risk of blockage. Some examples of alternative methods to produce higher turbulence are the modules with rotor and stator system, e.g. VRM-Modul in WWTP Knautnaundorf (Stein 2003).

Table 12.5 Advantages and disadvantages of dead-end (unsteady and submerged mode) and cross-flow mode in membrane filtration of wastewater (Chang and Fane, 2000; ATV-DVWK 2002b; Melin and Rautenbach 2004).

Configuration of MBR	Advantages and disadvantages
Dead-end mode	Unsteady state process Requires low suspended solids in feed stream Low specific energy costs of 0.1–0.5 kWh m ⁻³ Module and membrane blocking Back-washing if stable versus compression Long filtration intervals Periodic cleaning management Final filtration of wastewater
Submerged mode	Quasi-steady state High suspended solids in the feed treatable Low specific energy costs of 0.3–0.7 kWh m ⁻³ Low fluxes of about 10–30 L m ⁻² h ⁻¹ Less stress for biomass Two-phase flow pattern for hydrodynamical influence on deposits, permanent bubbling High investment, low operational costs Back-washing with and without air Short filtration intervals Periodic cleaning management
Cross-flow mode in steady state	Requires reversible gel layer for steady state process High suspended solids treatable Highest specific energy costs of 2.5–6.0 kWh m $^{-3}$ High fluxes of about 100 L m $^{-2}$ h $^{-1}$ With cross-flow of $w_{\rm cf}$ 3–6 m s $^{-1}$ High stress for biomass Low investment but high operational costs Back-washing with and without air Periodic cleaning management

Another way to control gel layer formation is to use interval back flushing but with the disadvantage of a poor water recovery rate in the range of 80-90% of the feed flow. Intervals for back-flushing and chemically enhanced cleaning depend greatly on water quality and are often necessary in the case of river water processing (Dietze 2004).

12.4.3

Membrane Fouling and Cleaning Management

12.4.3.1 Types of Fouling Processes

The major symptoms of fouling are a decline in flux over operating time, increasing transmembrane pressure, sludge cake formation and changes to the retention coefficient R_t (see Eq. 12.75). Stationary filtration behavior is not possible. Fouling occurs if a critical flux is exceeded. Chang et al. (2002) defined the critical flux as the highest flux for which the transmembrane pressure remains constant based on data from a step-by-step increase in flux, depending on the type of membrane material, MLSS and cross-flow velocity. However, there is a transition between concentration polarization and stagnant cake formation (Chen et al. 1997). For a relatively high c₃, polymerization and precipitation can occur, resulting in solid cakes. To a certain extent, nearly every feed component leads to membrane fouling.

The formation of surface deposits on membranes is influenced by feed composition, flow conditions, the chemical nature of the membrane and interactions between components and the membrane. The type of fouling depends on the nature of deposit components (Fleming 1995). We distinguish:

- Scaling or mineral fouling is the deposit of inorganic material with crystal structures like salts.
- Organic fouling is the deposit of organic material like grease, oil, surfactants, proteins and humic substances.
- Colloidal fouling is the deposit of particles like clay and metal-oxides or hydrox-
- Bio-fouling is the formation of bio-films by microorganisms captured and growing at the surface. A secondary phenomenon is the excretion of enzymes and extracellular polymeric substances (EPS) which influence the gel layer.

Deposits which can be detached by cleaning processes, like back-flushing and mechanical cleaning, are called reversible fouling and cause a reversible gel layer formation. Deposits which lead to an irreversible gel layer formation can only be removed by chemical cleaning procedures and are called irreversible fouling (Rautenbach and Albrecht 1989). Therefore, fouling can be controlled only to a certain degree by hydrodynamics.

Different fouling mechanisms are shown in Fig. 12.9. An irreversible sludge cake layer is formed by particles, contaminants and agglomerates of contaminants which are bigger than the pore size of the membrane.

Due to the heterogeneous nature of bioreactors, mixed liquor fouling is difficult to predict and control in a MBR. Factors affecting fouling are (Chang et al. 2002; Lee et al. 2003; Rosenberger 2003; Shon et al. 2004):

- The membrane material hydrophobicity, porosity and pore size and distribution.
- The mass of microorganism MLSS and of extra-cellular polymeric substances EPS, floc structure, dissolved matter and floc size.
- The operating conditions e.g. configuration, cross flow velocity, aeration, hydraulic and solid retention time and trans-membrane pressure.

12.4.3.2 Membrane Cleaning Strategies

To reduce the two negative influences of concentration polarization and decreasing flux rates which result from fouling, a cleaning strategy has to be developed together with the membrane suppliers which is adapted to the wastewater characteristics, the membrane material and the configuration of the membrane filtration

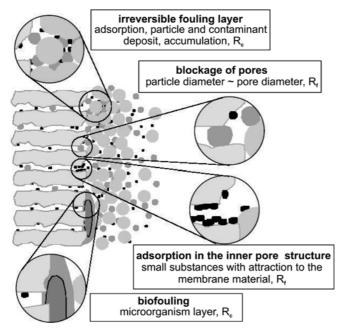


Fig. 12.9 Fouling mechanisms, adapted from Melin and Rautenbach (2004).

process. In general, process integrated back-flushing during the filtration process is used to maintain a constant flux. Periodically *in situ* cleaning with chemicals and intensive washing with chemicals *ex situ* are necessary to remove irreversible deposits.

MBR processes in submerged or cross-flow mode often use fixed-interval backwashing or back-flushing with permeate, whereby reverse flow is used to wash-out reversible fouling. Usually, a filtration process cycle of about 10 min is followed by a back-washing time of 1 min, resulting in a water recovery of at least 90% as shown in Fig. 12.8. Experiments are necessary to optimize two factors: (a) permeate loss by back-washing and (b) the slight decrease in permeate flux which requires maintenance cleaning to recover 100% flux. In submerged systems, intensified aeration in a range of $0.21-3.0 \text{ m}^3 \text{ m}^{-2} \text{ h}^{-1}$ is also used to prevent fouling (Benedek and Côté 2003; Cornel et al. 2001).

Maintenance cleaning is done at longer intervals with various chemicals. The chemicals used and the interval chosen must be specifically adapted to the range of application to avoid membrane damage or the formation of harmful substances (ATV-DVWK 2002b). Chemical cleaning usually restores the permeate flux but produces a certain amount of contaminated water.

In situ chemically enhanced cleaning procedures (intermediate cleaning) are used in MBR processes if the flux decreases by about 20% down to 100 L $\rm m^{-2}~h^{-1}$ bar $^{-1}$, or as dictated by a cleaning management time table.

Acidic solutions are used particularly for the elimination of inorganic fouling causing substances and alkaline solutions for the oxidation of organic foulants. Disinfectants may be used to eliminate microorganisms. Commonly used commercial purifiers contain active chlorine concentrations between 300 mg L⁻¹ (Wehrle Umwelt 2005) and 2000 mg L⁻¹ (MUNLV 2003), or may contain special mixtures of acids, leaching agents, surfactants, active enzymes (Berghof 2005) or hydrogen peroxide (AV Aggerwasser 2005). Tests are also often performed with citric acid, NaOH, HCL, oxalic acid (Wozniak 2003) and nitro-hydrochloric acid. Sodium hypochlorite is known to recover total permeability, but its use is decreasing because of the damage caused to microorganisms at high concentrations (MUNLV 2003). Heating of the purifiers, sometimes in a separate tank, may be necessary to reach the required cleaning temperature of 35-40°C.

An intensive ex situ cleaning (main cleaning) is necessary at least twice a year. The typical cleaning management program combines alternating chemical cleaning and clean water rinsing for neutralization and a reaction procedure enhanced with chemicals.

12.5 **Membrane Bioreactors**

12.5.1

Final Treatment (Behind the Secondary Clarifier)

The methods of industrial and municipal wastewater treatment have already achieved a high standard by combining biological and mechanical processes and additional treatment steps for the elimination of phosphorus and nitrogen (see Chapter 10). However, the microbiological quality of effluents is still a burden for surface waters. Pathogens like bacteria, viruses and parasites are a problem, particularly for bathing waters in summer, or in areas with drinking water reservoirs. There are different standards in effect, such as the EU Water Framework Directive (EU 2000) with the aim to reach a high ecological water quality by 2015, or the EEC Council Directive 76/160/EEC of 8 December 1975 concerning the quality of bathing waters (EEC 1991; Table 12.6).

Ultra- and microfiltration are of high interest as a post-treatment method after sedimentation, and recently, as an alternative to the traditional gravity settlers used in municipal wastewater treatment to meet the quality standards mentioned above.

The high efficiency of membrane systems as a final treatment in municipal WWTP was investigated and proven by Altmann et al. (1995) in Berlin-Ruhleben. In pilot-scale experiments, five different membrane systems were investigated in combination with phosphate precipitation. Microorganisms were found to be eliminated by a factor of several orders of magnitude and viruses were typically adsorbed on suspended solids, which were eliminated (see Fig. 12.10 in Section 12.5.2). An alternative is to couple biological treatment with sand filtration.

Table 12.6	Requirements for post treatment of effluents from WWTP
Microbiolo	gical Standard in EEC Council Directive 76/160/EEC of
8 December	er 1975 concerning the quality of bathing water (EEC 1991).

Parameter	Imperative value	Guide value	
Total coliform bacteria (cells/100 mL)	500	10 000	
Fecal coliform bacteria (cells/100 mL)	100	2 000	
Streptococcus faecalis (cells/100 mL)	100	-	
Salmonella (cells/1 L)	-	0	
Bowel viruses (PFU/10 L)	-	0	

12.5.2 Membrane Bioreactors in Aerobic Wastewater Treatment

The combination of biological and membrane filtration processes is known as a membrane bioreactor (MBR) system. The benefits of MBR systems in submerged or external cross-flow mode are (ATV-DVWK, 2000b; DWA 2005):

- High microbiological quality of effluent water resulting from the high removal efficiency of suspended solids, microorganisms and viruses.
- Greater freedom to vary process parameters, like the reduction of excess sludge or the concentration of slowly growing organisms because MBR systems are independent on the sedimentation behavior of sludge.
- Volume reduction of the activated sludge tank because of higher biomass concentration and elimination of the sedimentation tank, both resulting in a reduction of the total plant footprint (Côté and Liu 2003).

The financial success of MBR is largely determined by the processes used to restore decreased flux caused by fouling. The resulting disadvantages of MBR are the high investment cost for the needed membrane surface area and/or high operating costs for cleaning management and the energy demand for cross-flow mode on top of the cost for supplying oxygen to microorganisms (Choi 2005; DWA 2005).

The discharge of harmful substances, like endocrine-disrupting substances (EDS) into municipal and industrial wastewater and their passage through WWTP has also been investigated (Filali-Meknassi et al. 2004). Reverse osmosis and nanofiltration processes are able to reject EDS (Fig. 12.1), but sorption was found to be the main elimination process in sewage and industrial wastewater treatment, hence the elimination efficiency was high for MBR (Kunst 2002; Gallenkemper et al. 2003; Schäfer et al. 2003; Oschmann et al. 2005).

Recently, submerged membranes have been applied in large-scale activated sludge plants more and more often because of the economical advantages of low pressure processes. By 2004, several commercial municipal WWTP in Germany had been built with submerged MBR systems for more than 100 000 inhabitants; and common permeate fluxes are 8-30 L (m² h)⁻¹. The percentage of submerged systems is still increasing, evidently for small-scale systems (Bischof et al. 2005; Brinkmeyer et al. 2005).

Figure 12.10 gives an overview of the membrane filtration process in comparison with conventional wastewater treatment.

In cross-flow mode (Fig. 12.10c), the higher operational pressure of about 3 bar is provided by pressure pumps and a loop or recycling stream is used to increase shear at the membrane surface to attain high fluxes (e.g. BIOMEMBRAT used in activated sludge plants of food industries). Air bubbling and periodic back-flushing is possible. To date, no application of cross-flow mode in municipal WWTPs has been realized.

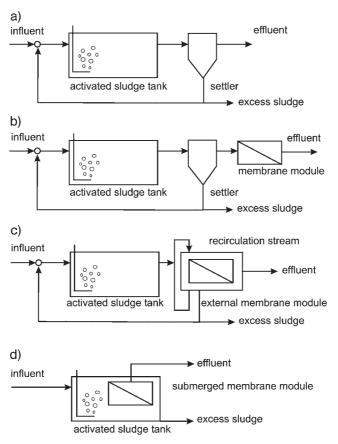


Fig. 12.10 MBR systems, external and submerged mode in comparison with a conventional activated sludge system.

Table 12.7	Performance characteristic of conventional activated sludge
process an	d MBR (Günder 1999; ATV-DVWK 2000b; Cornel et al. 2001;
Côté and L	iu 2003; MUNLV 2003).

Parameter		Conventional WWTP	MBR
X	g L ⁻¹ MLSS	<5	8.0–16.0
t_{RX}	d	15	15.0
t_R	h	23	3.6-8.0
B_x	kg BSB ₅ kg ⁻¹ d ⁻¹ MLSS	< 0.08	< 0.08
α , O_2 transfer ratio	-	0.6	0.6
Effluent data			
S	mg L ⁻¹ COD	40-50	<30.0
$N_{\rm ges}$	mg L ⁻¹	<13	<13.0
P_{ges}	mg L ⁻¹	0.8-1.0	< 0.3
X _e	mg L ⁻¹ MLSS	10-15	0
Microbiological	_	Hygienically	Bathing
quality		critical	water quality

In the submerged mode (Fig. 12.10d), the membrane modules are directly installed in the activated sludge bioreactor or immersed in an aerated separate container (e.g. ZeeWeed-Cassettes; Cornel et al. 2001). Air bubbling plays a significant role in attaining high fluxes (Section 12.4.3) and periodic back-flushing is used to reduce fouling.

The retention efficiency of the separation process is not influenced by gas bubbles, floating sludge or bulky flocs with lower density. Performance characteristics of MBR systems are given in Table 12.7 in comparison with conventional WWTP. The minimum solid retention time with nitrification is 15 d for both systems, according to ATV-DVWK (2000a).

In the case of a MBR, the retention time of the sludge is unaffected by sedimentation behavior (Fig. 12.10c, d) and very high sludge ages (see Section 6.2.3) can be reached; but for municipal WWTP sludge ages of 15 d and sludge concentration of 8–16 g L⁻¹ MLSS are common (Table 12.8). Activated sludge with such high concentration exhibits non-Newtonian behavior, the apparent viscosity is a function of g L⁻¹ MLSS as well as shear gradient and affects both oxygen mass transfer and the degree of mixing (Rosenberger 2003; Kubin 2004; Choi 2005). The relative oxygen transfer ratio α_w (see Eq. 5.12) for different WWTP under operational conditions is found to be in the range of 0.25 to 0.8 for X and the range of 1–17 g L⁻¹ MLSS (Cornel et al. 2001; Drews and Kraume 2005; DWA 2005).

Removal efficiency rises with increasing MLSS concentration, but a maximum removal of 96–97% cannot be exceeded. COD removal efficiency was summarized by Kubin (2004) and Drews and Kraume (2005).

We summarize data for some German MBRs in the field of municipal wastewater treatment in Table 12.8. These all operate with submerged membrane modules. The positive experience gained over several years of operation has led to fur-

Table 12.8 Membrane bioreactors in municipal wastewater treatment plants, submerged mode (Engelhardt 2003; Stein 2003; Voßenkaul et al. 2003; MUNLV 2003; Wozniak 2003; De Wilde et al. 2005; Roest et al. 2005; Rondi and Montagnoli 2005).

Year	WWTP operator	Company, membrane, configuration, submerged mode	Q m³h ⁻¹)	A _M (10 ³ m ²)
1999	Rödingen Erftverband	Zenon, hollow fiber	135	4.846
2004	Nordkanal Erftverband	Zenon, hollow fiber	1881	85
1999	Büchel Aggerverband	Kubota, plate membrane	40	0.96
2004	Seelscheid Aggerverband	Kubota, plate membrane	356	12.48
2000	Markranstädt kommunale Wasserwerke Leipzig	Zenon, hollow fiber	180	8.8
2002	Knautnaundorf kommunale Wasserwerke Leipzig	Martin System AG, rotating plate membrane	23	0.756
2004	Markkleeberg kommunale Wasserwerke Leipzig	Zenon, hollow fiber	710	28.4
2003	Simmerath Wasserverbd. Eifel-Ruhr	Puron, hollow fiber	Bypass	1.0
2003	Monheim Stadt Monheim	Zenon, hollow fiber	288	12.32
2004	Waldmössingen Stadtwerke Schramberg	Zenon, hollow fiber	90	5.3
2003	Schilde (Belgium)	Zenon, hollow fiber	355	10.56
2003	Brescia (Italy)	Zenon, hollow fiber	1750	-
2004	Varsseveld (The Netherlands)	Zenon, hollow fiber	755	20.16

ther applications. For example, in WWTP Markranstädt (Stein 2003) the hollow fiber membrane filtration modules are located in the upper part of the nitrification tank. Mixing units are installed together with aeration to reduce fouling. An *ex situ* cleaning shaft located between the nitrification lines is in place to take up the modules for the cleaning procedures. Another concept is followed with separated membrane filtration containers in WWTP Büchel (Wozniak 2003) and WWTP Rödingen (Engelhardt 2003). These filtration containers are aerated and the wastewater is recycled between the nitrification and denitrification tanks and the filtration containers, where nitrification also takes place.

Experience with the WWTPs mentioned above has shown the importance of an intensive pre-treatment of wastewater by filtration. To reduce membrane fouling by the deposit of particles, it is necessary to perform additional pre-treatment of wastewater with a grid or a sieve <1 mm (Engelhardt 2003; Stein 2003; DWA 2005). Especially the fibrous components of the untreated wastewater leave deposits in the upper part of hollow fiber modules.

The costs associated with MBR systems operating in cross-flow and submerged mode have been analyzed by Melin and Rautenbach (2004), who showed in estimated overall operating cost of 0.57 € m⁻³ for cross-flow mode and 0.56 € m⁻³ for submerged mode in sewage WWTP at small scales with volume rates of 100 m³ h⁻¹. Overall operational costs for existing MBRs in wastewater treatment in the food industries were found to be 0.90 € m⁻³ (Wienands and Streif 2005), while the highest costs of cross-flow mode come from the energy consumption and the highest costs for submerged mode result from the investment in the membrane. The energy consumption of submerged systems has been measured between 0.8 kWh m⁻³ and 4 kWh m⁻³. Optimization of the aeration management leads to a reduction of energy consumption by about 35% and a consumption of less than 1 kWh m⁻³ has been predicted (Engelhardt 2003).

12.5.3

Membrane Bioreactors and Nutrient Removal

To achieve nitrogen removal with a MBR system, aerobic-anoxic operation conditions are necessary as in the conventional process (see Section 10.4.2). A MBR offers good nitrification conditions because of the high retention of sludge allowed. Therefore, even slowly growing nitrifying bacteria have an excellent chance to establish themselves in the aerated tank. Nitrogen removal with preliminary denitrification (see Fig. 10.9a) and with post-denitrification (see Fig. 12.11b) is in operation with MBRs (Brepols et al. 2005). Investigations have also been performed in small-scale applications with post-denitrification without dosage of a carbon source (Gnirß 2005; Maas 2005).

The high oxygen transfer associated with submerged membranes creates a need for a larger denitrification zone (Engelhardt 2003; Gnirß et al. 2003; MUNLV 2003). In the case of post-denitrification, the membrane modules are located in a separate container behind the anoxic zone with optiomal aeration to avoid sludge cake formation (Adam 2004; Drews et al. 2005).

Phosphorus elimination by simultaneous precipitation is a well known technique in conventional wastewater treatment and is also used in MBR. The high MLSS retention efficiency of the membrane process significantly reduces the discharge of coagulant chemicals and leads, therefore, to less loading of rivers. In surface water processing, Dietze (2004) achieved low phosphate concentrations (<15 μg L⁻¹ PO₄-P) in permeate using phosphorus flocculation and membrane filtration while reducing the coagulant and salts discharge, compared to traditional flocculation and sand filtration.

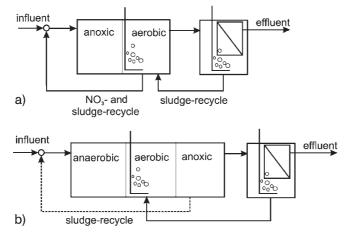


Fig. 12.11 MBR and nutrient removal with submerged membranes with: (a) preliminary denitrification, (b) post-denitrification without substrate dosage and with biological phosphorus removel.

Investigations have been conducted to combine biological phosphorus elimination with an anaerobic zone, not only in WWTP (Section 10.4.3, Fig. 10.9a) but also in the MBR process to reduce the use of chemicals like FeCl₃. The question remains whether the high sludge ages used in MBR yield higher phosphorus elimination capacities. The high retention of MLSS and the prevention of phosphorus release caused by the high oxygen concentration in the MBR system lead to a high efficiency (Gnirß et al. 2003). Orthophosphates from release cannot be retained by ultrafiltration membranes (Adam 2004).

PROBLEM 12.1

When do we have to consider the impact of osmotic pressure? Calculate the osmotic pressure for two aqueous solutions, considering complete retention (c_2 =0 mg L^{-1}) and neglecting concentration polarization. The feed contains 15 g L^{-1} NaCl and 15 g L^{-1} glucose (T = 288.2 K, R = 8.31 J mol⁻¹ K^{-1}).

Solution

NaCl, molar mass $M=58~g~mol^{-1}$, the osmotic pressure increases as a result of dissociation as the number of moles increases (Rautenbach and Albrecht 1981). For complete dissociation of NaCl $\beta=2$ follows, we obtain from Eq. (12.24b):

$$\Delta \Pi = \frac{2 \cdot 15 \cdot 8.31 \cdot 288.2}{58} \frac{\text{g J mol K}}{\text{L g mol K}} = 12.4 \frac{10^5 \text{ J}}{\text{m}^3} = 12.4 \frac{10^5 \text{ N}}{\text{m}^2} = 12.4 \text{ bar}$$

Glucose, molar mass M = 181 g mol⁻¹. At the cut-off of nanofiltration membranes, we obtain from Eq. (12.24a):

$$\Delta \Pi = \frac{15 \cdot 8.31 \cdot 288.2}{181} \frac{\text{g J mol K}}{\text{L g mol K}} = 198 \frac{\text{J}}{\text{L}} = 2 \frac{10^5 \text{ J}}{\text{m}^3} = 2 \frac{10^5 \text{ N}}{\text{m}^2} = 2 \text{ bar}$$

Considering concentration polarization and increasing concentration of the feed in the direction of flow, the osmotic pressure increases. For particles like bacteria or colloids >10000 g mol⁻¹, the influence of osmotic pressure is negligible.

PROBLEM 12.2

What membrane surface area is required for a WWTP for 100000 inhabitants, considering permeate fluxes J_{p10} at 10 °C? ($J_{p8} = 25 \text{ L m}^{-2} \text{ h}^{-1}$)

The common design method of a WWTP is based on ATV-DVWK-Arbeitsblatt A 131 (ATV-DVWK 2000a). The maximum flow rate Q_m is determined by the dry-weather flow Q_s and the average wastewater from other areas Q_f. According to ATV-DVWK-Arbeitsblatt A 198 (ATV-DVWK 1992), the flow rate is given by:

$$Q_{\rm m} \ge 2 Q_{\rm s} + Q_{\rm f} L \, {\rm s}^{-1} \tag{12.79}$$

To determine the maximum inlet flow rate Q_m we choose:

$$Q_{\rm m} = 3.5 \, Q_{\rm s} + Q_{\rm f} \tag{12.80}$$

Q_s is the average sewage flow rate according to the median discharge per household, commercial and industrial wastewater, here for 100000 inhabitants, with a specific volume rate of inhabitants (inh.) of 130 L (inh. d) $^{-1}$.

Q_f is annual average extrameous wastewater from other areas, here we choose 6000 m³ d⁻¹.

Solution

For $100\,000$ inhabitants, $Q_{\rm m}$ is given by:

$$Q_{\rm m} = \frac{3.5 \cdot 130 \cdot 100\,000 \cdot 10^{-3} + 6000}{24} \; \frac{L \; inh. \; d}{inh. \; d \; h} \; \cdot \; \frac{m^3 \; d}{d \; h} = 2146 \; m^3 \; h^{-1}$$

while the average daily flow rate Q_d (dry-weather flow) is:

$$Q_d = Q_s + Q_f \tag{12.81}$$

The required permeate flow rate of a membrane module has to equal the maximum volume rate Q_m ; and the required membrane surface area A_M can be determined from permeate flux of common membrane modules. At 8 °C, a flux of $J_{p8} = 25 \text{ L m}^{-2} \text{ h}^{-1}$ is given. For higher temperatures (here 10 °C) the flux is about 15% higher than at 8°C (MUNLV 2003), resulting in:

$$J_{\rm p10} = 1.15 \cdot 25 \text{ L m}^{-2} \text{ h}^{-1} = 28.75 \text{ L m}^{-2} \text{ h}^{-1}$$

According to Eq. (12.5), the required membrane surface area $A_{\mbox{\tiny M}}$ follows:

$$A_{\rm m} = \frac{Q_{\rm m}}{J_{\rm p10}} = \frac{2146}{28.75} \cdot \frac{\rm m^3 \, m^2 \, h}{\rm L \, h} = 74\,638 \, \rm m^2$$

For maintenance cleaning twice a year, the complete membrane area is taken out of operation. Each cleaning procedure lasts about 1 day; the effective membrane area is in operation, therefore, only 363 days year⁻¹ or about 99% of the time. To realize the required flux, the total membrane surface area A_t must be 101% of A_M .

$$A_t = A_M + 0.01 \ A_M = 75.4 \cdot 10^3 \ m^2$$

PROBLEM 12.3

How different are the volumes of a MBR and an activated sludge tank, for the same treatment task with nitrification?

Conditions with full nitrification require sludge ages of more than 15 days (ATV-DVWK 2000). The common method for a single-step activated sludge process is based on the sludge loading rate B_x (Günder 1999):

$$B_{\rm X} = \frac{{\rm S} \ {\rm Q_d}}{{\rm V} \ {\rm X}} \ {\rm kg} \ {\rm BOD_5} \ ({\rm kg} \ {\rm MLSS} \ {\rm d})^{-1}$$
 (12.82)

where B_x is the sludge loading rate, Q_d is the dry-weather flow rate according to Eq. (12.81) = $19\,000$ m³ d⁻¹, X is the concentration of bacteria and S is the concentration of substrate (we choose 400 mg L⁻¹ BOD₅). For the MBR we choose $B_X = 0.03$ kg BOD_5 (kg MLSS · d)⁻¹ and for a conventional plant a higher loading rate of 0.06 kg BOD₅ (kg MLSS \cdot d)⁻¹.

Solution

Under the assumptions of no oxygen limitation and identical percentage of living bacteria, an approach for processes with and without membrane is given by:

1. With the membrane, the concentration of microorganisms is in the range of 8–16 g L^{-1} MLSS. We choose X = 12 g L^{-1} MLSS (Table 12.7). The volume of the aerated sludge tank for MBR follows from Eq. (12.82):

$$V = \frac{S~Q_{\rm d}}{X~B_{\rm X}} = \frac{0.4 \cdot 19\,000}{12 \cdot 0.03} ~\frac{m^3~kg~m^3~kg~d}{kg~m^3~d~kg} = 21.1 \cdot 10^3~m^3$$

2. In the conventional activated sludge process under the same conditions of S and Q_d , the attainable concentration of microorganisms is $X = 3 \text{ g L}^{-1}$ MLSS and a higher sludge loading rate of 0.06 kg BOD₅ (kg MLSS d)⁻¹ (Table 12.7) is possible. The volume of the single-step activated sludge tank is:

$$V = \frac{S~Q_{\rm d}}{X~B_{\rm X}} = \frac{0.4 \cdot 19\,000}{3 \cdot 0.06} ~\frac{m^3~kg~m^3~kg~d}{kg~m^3~d~kg} = 42.2 \cdot 10^3~m^3$$

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