Process Improvement by Application of Membrane Bioreactors

A. Drews, M. Kraume
TU Berlin, Institut für Verfahrenstechnik, Str. des 17. Juni 136, 10623 Berlin, Germany;
tel. +49 30 314 23701, fax +49 30 314 21134, e-mail: anja.drews@tu-berlin.de.

1 Introduction

In cultivating microorganisms for either wastewater treatment or biotechnological production, high biomass concentrations are desirable to increase removal rates or volumetric productivity. Separating the biomass from the liquid (product) phase can however become problematic at higher biomass concentrations. Therefore, in wastewater treatment, biomass concentrations of only 4-5 kgMLSS m^{-3} are applied in the common activated sludge process (ASP). At higher concentrations, enormous sedimentation tanks would be required to settle sludge while floating sludge would be even impossible to settle. In fermentations, often fed-batch processes are applied to achieve high biomass concentrations. If a continuous process is sought, utilising a chemostat process would waste large amounts of productive biomass and raw materials. In both cases, it is advantageous to use a membrane bioreactor (MBR) which combines the benefits of high biomass concentrations with the possibility to run a continuous process at controlled biomass retention. Since membrane costs have decreased dramatically over the last couple of years (to approx. 50 €/m² nowadays (MUNLV, 2003)), and energy requirements for aeration of the membrane are also fast approaching the normal ASP range, this has now become an economically feasible solution even for low-profit processes such as wastewater treatment. Here, application of a membrane does not only reduce the required reactor volume but also makes sand filtration and disinfection redundant as it produces a hygienic effluent (see Fig. 1)

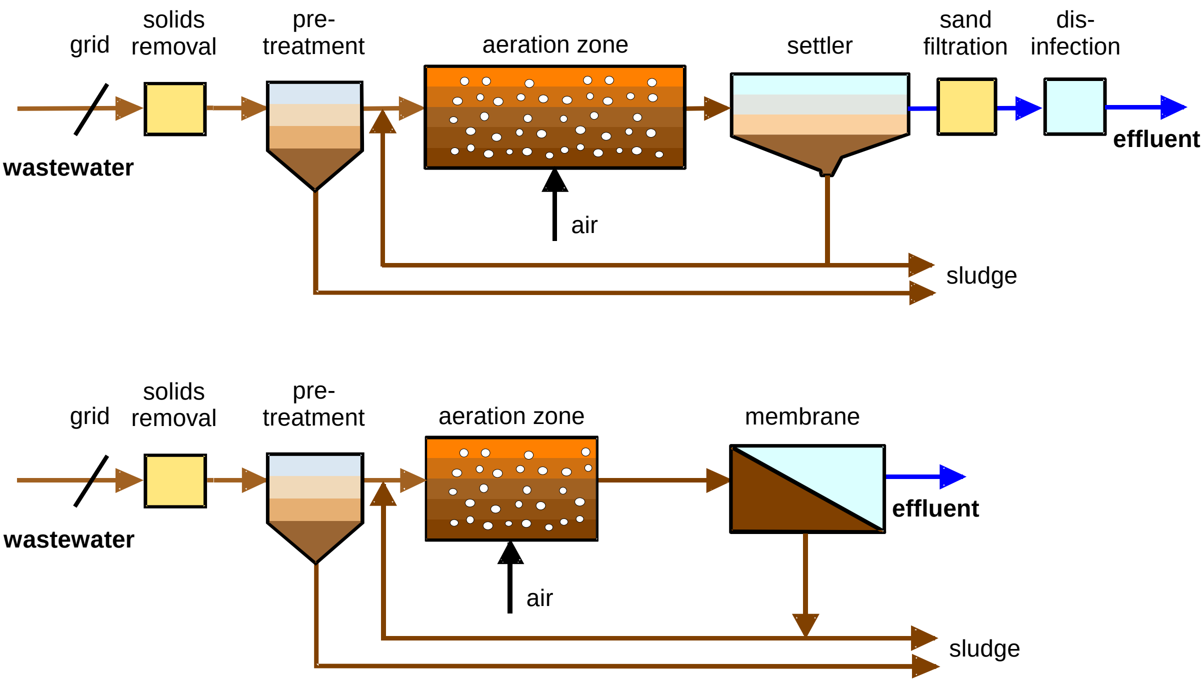


Fig. 1: Wastewater treatment plant set ups: common activated sludge process (ASP, top) and membrane bioreactor (MBR, bottom).

2 What is a membrane bioreactor?

Membrane bioreactors (MBR) are combinations of common bioreactors and membrane separation units for biomass retention. Commonly, micro- or ultrafiltration membranes with pore sizes ranging from 0.01 – 0.4 µm are applied. The membrane module can either be immersed in the biosuspension or set up in the sidestream mode (see Fig. 2).

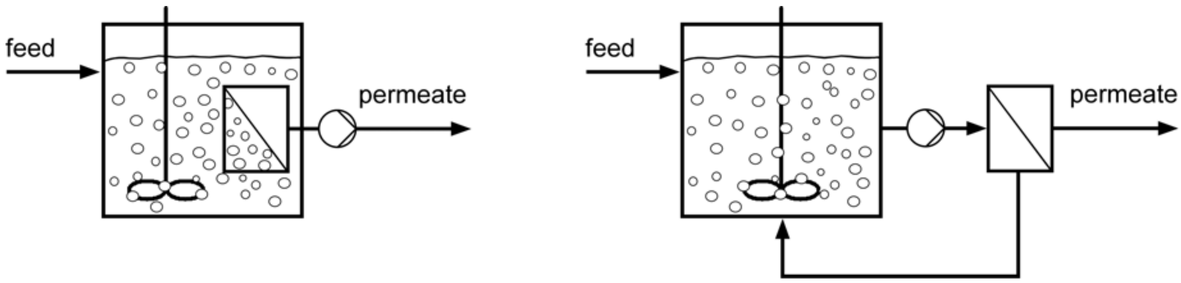


Fig. 2: MBR set ups: immersed (left) and sidestream (right)

Membrane bioreactors (MBR) allow significant process intensifications and better effluent qualities due to the following changes of boundary conditions:

- MBRs are operated at higher biomass concentrations with resulting high metabolic rates. This allows decreased reactor volumes and footprint.
- Since hydraulic and solids (biomass) residence times are independent of each other, MBRs offer an additional degree of freedom for process control. Therefore, they can be particularly advantageous in fermentations where inhibitory metabolites occur either as the desired product or as unwanted byproducts since the productive cells are retained in the vessel while soluble products are continuously withdrawn. In wastewater treatment, degradation kinetics can also be optimised beyond ASP performance as even slowly growing microorganisms with particular degradation features can be established.
- Since no gravitation settler is needed, operation is independent of sludge parameters (floating sludge is not a problem).
- The membrane being a barrier for suspended solids, MBRs produce a more hygienic effluent.
- The utilisation of maintenance energy demands offers the possibility of decreased excess biomass formation. In wastewater treatment, excess sludge disposal accounts for 30 - 60 % of the total processing costs (Arnot and Howell, 2001; Low and Chase, 1999). Therefore, wastewater treatment plants (wwtp) should be designed and operated such that pollutants are diverted from biosynthesis to functions associated with non-growth activities.

First applications of MBRs in wastewater treatment date back to the early 70s. In the meantime, three generations of MBR wastewater treatment plants have been developed and an increasing number of technical plants is coming into operation (Kraume et al., 2004). Although several practical experiences and data are available for MBR processes there is still considerable optimisation potential. For pure cultures, MBRs have so far only gained importance in kinetic investigations (Tros et al., 1996; Müller and Babel, 1996; Menshutina et al., 2001; Drews, 2004).

Significant productivity increases have been reported for different fermentations (Arnot and Howell, 2001; Drews, 2004). As can be seen from Fig. 3, the volumetric productivity of a properly run MBR fermentation can be increased by a factor of 15 (at a 12-fold increase in biomass concentration) in comparison to the average value of repeated batches.

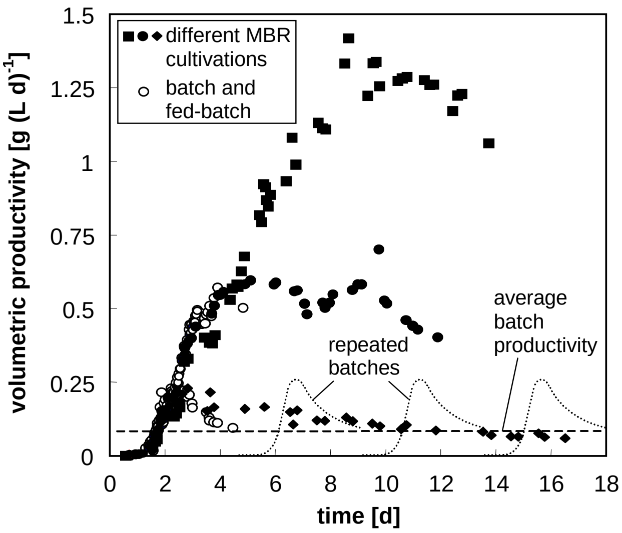


Fig. 3: Productivity increase (production of a siderophore by *Ustilago maydis* (Drews, 2004))

Fig. 4 shows the COD removal efficiency of different pilot and full scale MBR plants. At similar specific loading rates, removal increases with biomass concentration. This means that the cell specific removal rate increases. In a mixed population with different substrate uptake abilities this might be explained by the establishment of special organisms that are able to remove COD better or faster. A maximum removal of approx. 96 - 97 % cannot be exceeded as the remaining COD cannot be oxidised biologically. In this region, cell specific removal remains constant despite the fact that at lower specific growth rates (higher sludge ages) connected with higher biomass concentrations the specific substrate uptake rate of an organism decreases.

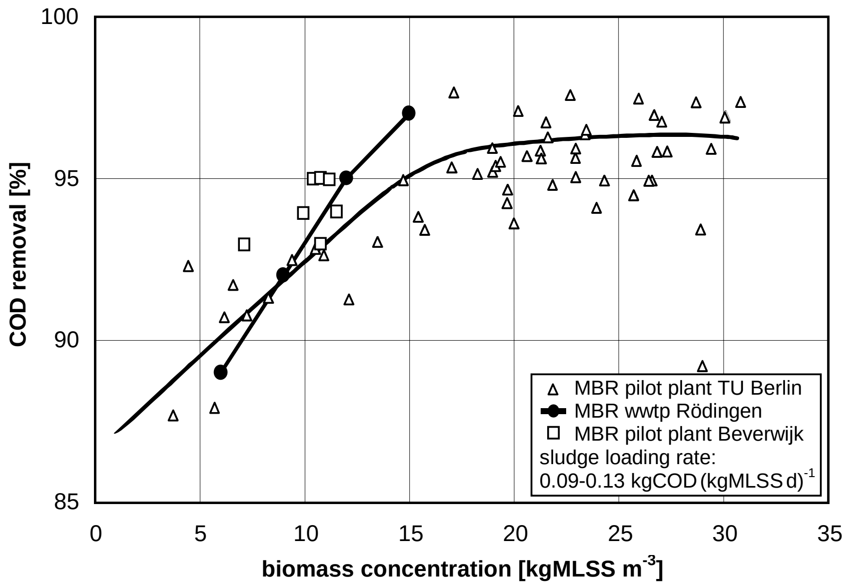


Fig. 4: COD removal efficieny of different pilot and full scale MBR in wastewater treatment (Kubin, 2004; Engelhardt et al., 2001)

Kinetics may also differ from ASP due to easier substrate access. In ASP, flocs may reach several 100 µm in size (Wisniewski et al., 1999). This means that the substrate can reach the active sites inside these flocs only by diffusion which causes an additional resistance and limits the overall reaction rate (diffusion controlled). Hydrodynamic stress in MBRs reduces floc size (to approx. 30 – 60 µm in immersed systems (Zhang et al., 1997; Song et al. 2003) and 3.5 µm in sidestream MBRs (Cicek et al., 1999)) and thereby indirectly increases the apparent reaction rate.

3 Why do MBRs have to be considered as hybrid reactors?

In the case of membrane bioreactors, 1 + 1 does not equal 2. Models and parameters describing biological reactions and membrane filtration have long since been in use. These, however, were determined in regimes distant from MBR regimes. Extrapolation of model structures and parameter values for use in MBRs is daring anyhow. In addition, the bioreactor and membrane stages cannot be regarded as individual unit operations since they interact with regards to both biological reactions and membrane filtration. These interactions, some of which have already been mentioned, are shown in Fig. 5. They will be explained in detail in the following chapter. Although several practical experiences and data are available for MBR processes no systematic investigation taking into account all interactions has been carried out so far due to the complexity of the system.

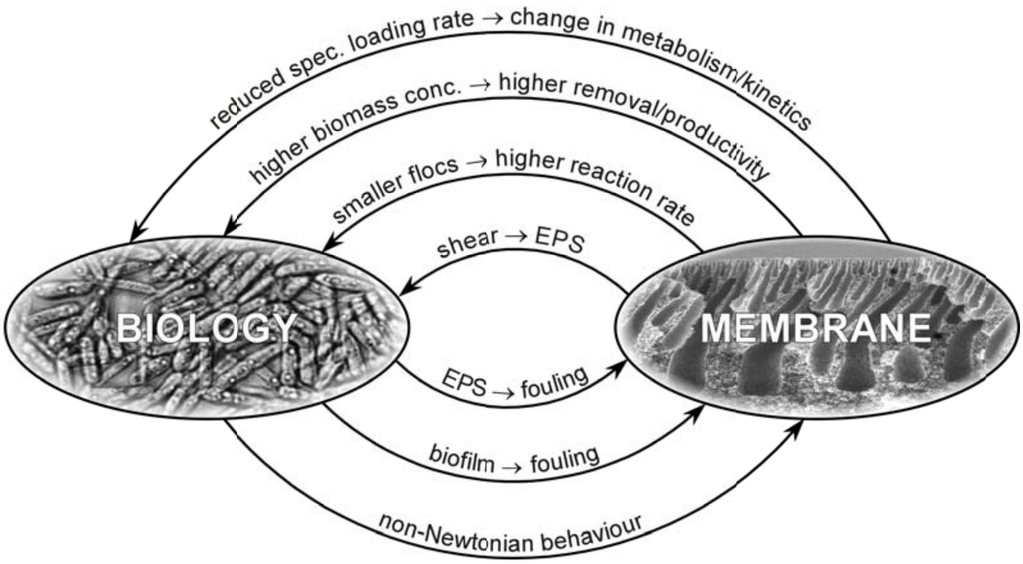


Fig. 5: Biology-membrane interactions

4 Experimental results

To discuss the effects of changed boundary conditions, regimes, and interactions, some experimental results on both biology and membrane filtration are presented in the following.

4.1 Biological process

4.1.1 Maintenance kinetics

Common models describing microbial growth and production or degradation do not consider the effect of high biomass concentrations or very low growth rates (i.e. high sludge ages) typical for MBRs and therefore cannot be applied. In this regime, maintenance metabolism where substrate uptake only yields energy for cell survival becomes of higher importance than in processes run at higher growth rates.

In degrading an organic substrate, carbon is directed mainly to formation of new cells and to CO_2 due to respiration. The relative yields of biomass and CO_2 depend on the physiological level of the biomass. At low growth rates, microorganisms utilise available substrates mainly for maintenance purposes. In wastewater treatment, this effect is observed as lower sludge yield $Y_{B/S}$ at high sludge ages (see Fig. 6) or at high biomass concentrations (Low and Chase, 1999), respectively.

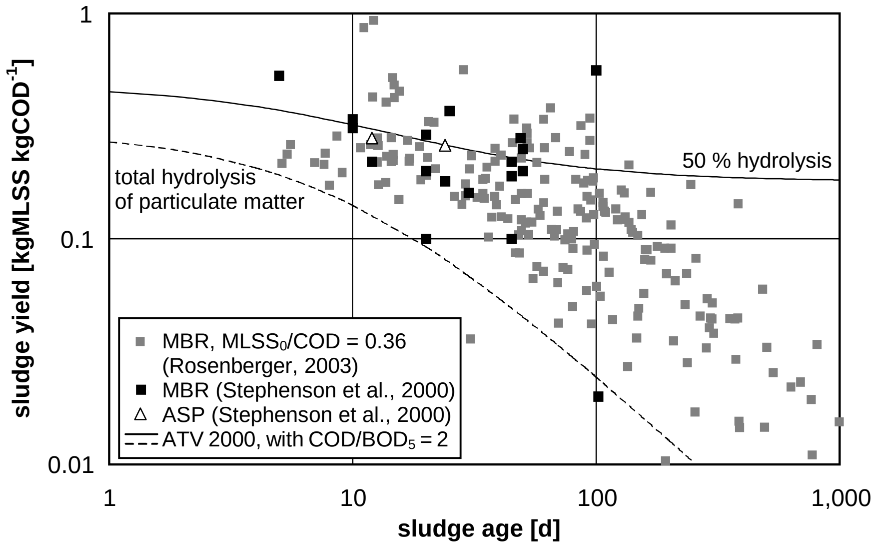


Fig. 6: Decrease of sludge yield at increasing sludge age in wastewater treatment. Curves represent model results based on total (---) and 50 % (—) hydrolysis of particulate matter in the influent (ATV, 2000).

Other processes which account for lower biomass yields but cannot be distinguished macroscopically are endogenous respiration or lysis and successive cryptic growth. Currently, when subjected to severe substrate deficiency, organisms are considered to fall into dormancy rather than die (van Loosdrecht and Henze, 1999). Only predation, adverse conditions or toxic substances or viruses lead to death. Whichever phenomenon prevails, most processes can be well described by Pirt's equation (1965), which in other words means $Y_{B/S} = f(\mu)$:

$$-\dot{r}_S = \frac{\dot{r}_B}{Y^g_{B/S}} + m_S \cdot c_B \qquad \text{or} \qquad \sigma = \frac{\mu}{Y^g_{B/S}} + m_S \qquad \text{(1) and (2)}$$

This linear relationship is shown in Fig. 7. Knowledge of the parameters maintenance coefficient m_S and true yield $Y^g_{B/S}$ permits modelling of the process including final biomass concentration c_B and substrate removal rate $-\dot{r}_S$. Considering literature data on aerobic and anaerobic growth of autotrophic and heterotrophic organisms in chemostats at temperatures ranging from 5 to 75 °C, Tijhuis et al. (1993) derived a thermodynamically based correlation for maintenance energy requirements. They concluded that $Y_{B/S}^{g}$ is constant at values of 0.45 – 0.5 kgCOD (kgCOD)$^{-1}$, with biomass concentration given here in kgCOD, too.

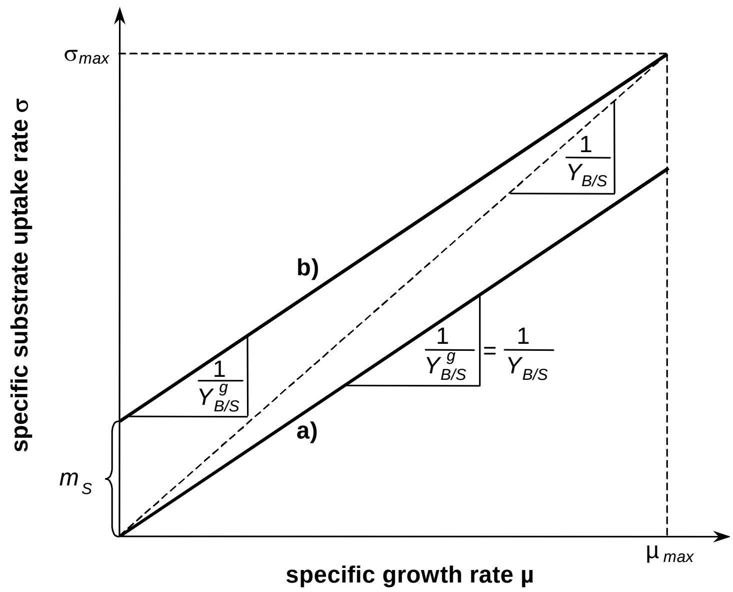


Fig. 7: Maintenance concept according to Pirt (1965): a) without maintenance, $Y_{B/S}$ = const, b) with maintenance, $Y_{B/S}$ = f(μ).

Depending on the range of operation, considering maintenance metabolism in growth models is of lesser or greater importance. At large growth rates, substrate uptake is dominated by growth purposes and m_S can be neglected. Since MBRs are operated at lower growth rates than ASP, ATV (German association for wastewater treatment) have proposed different parameters for modelling ASP and MBR processes (see Fig. 8).

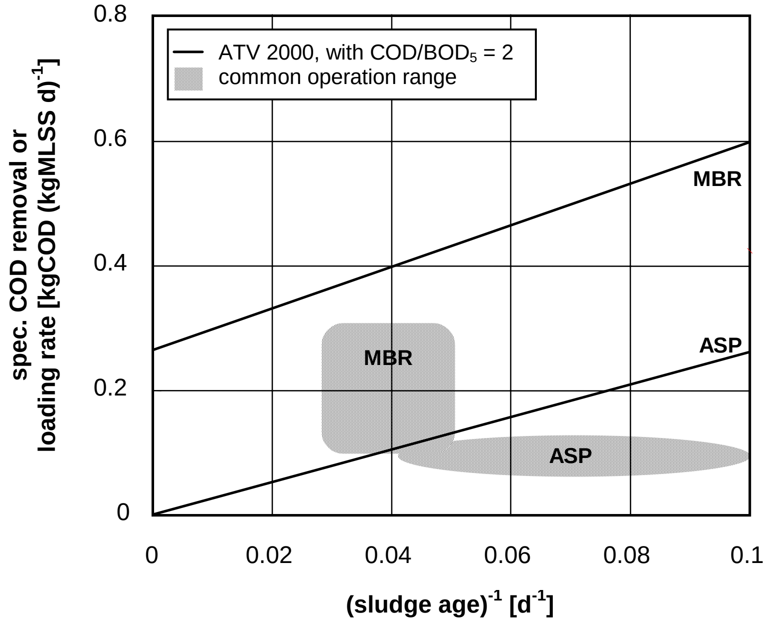


Fig. 8: Dependence of specific COD removal or loading rate on sludge age, common operation ranges (Kraume et al., 2004) and proposed parameters for ASP and MBR (ATV, 2000). (Under the assumption that COD removal is almost 100 %, removal rate equals loading rate.)

These, however, have not been determined in kinetic studies but have been fitted in the respective range of typical application. Commonly, microbial growth kinetics are determined in (fed-)batch or chemostat experiments. In MBRs, due to the rising biomass concentration each cell is subjected to an increasing substrate limitation which leads to a decrease in growth rate down to zero growth (see Fig. 9).

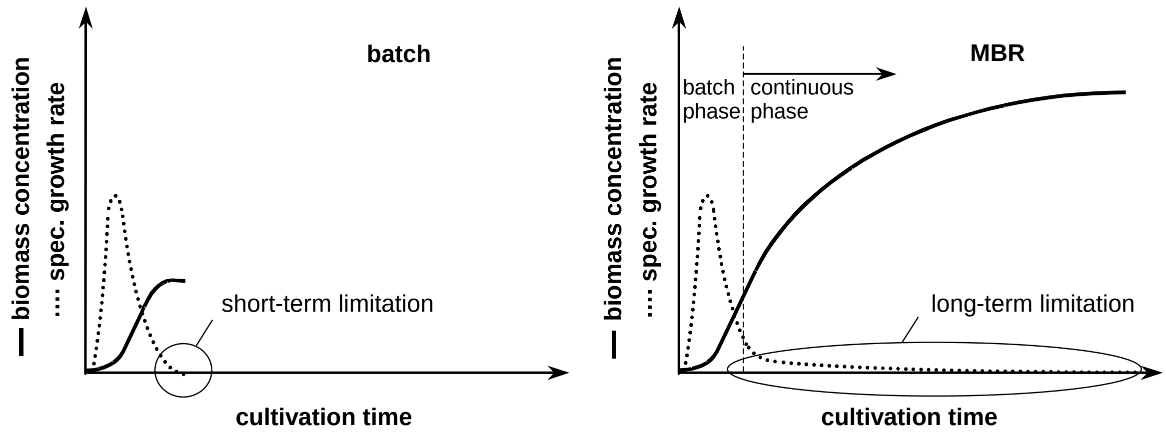


Fig. 9: Development of biomass concentration and specific growth rate in a batch and in an MBR cultivation.

Common models like the maintenance concept according to Pirt describe this as a progressive and continuous process, i.e. m_S = const. Anomalies of this concept have been reported for very low growth rates (< 10 % of μ_{max}), i.e. for severe substrate limitations caused by high biomass concentrations at a given loading rate typical for MBRs. At these very low growth rates or long-term limitations, microorganisms undergo changes in metabolism (Van Verseveld et al., 1984). Applying the Pirt concept, several authors have reported a significant reduction of maintenance demand at very low growth rates (Pirt, 1987; Low and Chase, 1999; Müller and Babel, 1996). In a study cultivating *P. fluorescens* on synthetic wastewater, however, Bouillot et al. (1990) found that neither $Y^g_{B/S}$ nor m_S were influenced by operating parameters such as dilution or breeding rate (even at < 5 % of μ_{max}). According to Wisniewski et al. (1999), hydraulic residence time in MBRs does not influence the maintenance coefficient, while Bulthuis et al. (1989) found a decrease of both m_S and $Y^g_{B/S}$ at lower residence times. While quite a few variations of m_S have been observed, no general correlation for its dependence on hydraulic residence time, growth rate or sludge age has been reported yet. For wastewater treatment MBRs, Bouillot et al. (1990) and Wisniewski et al. (1999) suggest $m_S \approx 0.04$ mgCOD (mgVSS h)$^{-1}$ and $Y^g_{B/S} \approx 0.36$ mgVSS (mgCOD)$^{-1}$.

Using an inaccurate value of m_S can lead to dramatic miscalculation of process data. As can be seen from Fig. 10, using $m_S = 0.027$ h^{-1} which was derived from short-term limited trials, the model fits measured fed-batch data well while it clearly underestimates biomass formation in an MBR cultivation. In the long run, this is well represented by using approx. 1/3 of the aforementioned value. In the initial unlimited phase, $m_S = 0.027$ h^{-1} yields a better fit.

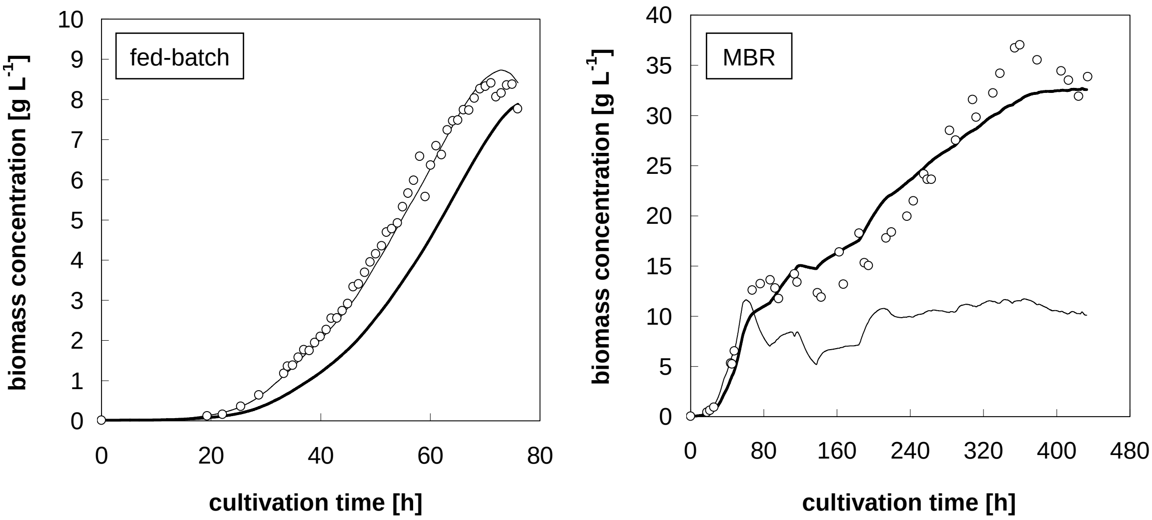


Fig. 10: Modelling of biomass concentration during fed-batch and MBR cultivation of *Ustilago maydis* using different values of the maintenance coefficient: — m_S = 0.008 h^{-1}, — m_S = 0.027 h^{-1} (Drews, 2004).

The minimum substrate concentration at which cells can just about duplicate also differs from that found by extrapolation of chemostat data (Tros et al., 1996). Assuming a Michaelis-Menten-type substrate uptake,

$$\sigma = \sigma_{max} \cdot \frac{c_S}{K_S + c_S} \tag{3}$$

yields the minimum substrate concentration at which $\sigma = m_S$:

$$c_{S,min} = \frac{K_S \cdot m_S}{\sigma_{max} - m_S} \tag{4}$$

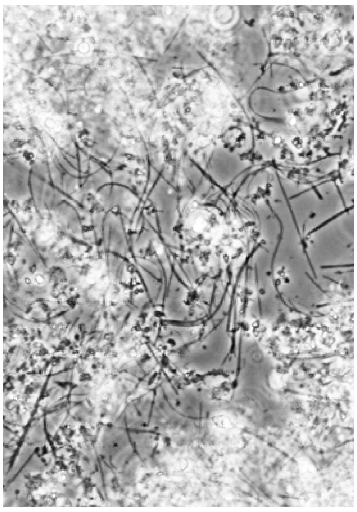
From eq. (4) it is apparent that $c_{S,min}$ decreases at lower values of m_S. Tros et al. (1996) determined a minimal acetate concentration of 26 µmol L^{-1} in an MBR in contrast to 109 µmol L^{-1} in a chemostat. The authors also found a decrease of the half-saturation constant down to a quarter of the value measured in the stationary phase of a batch run.

If a change in growth behaviour is existent at low growth rates, a change in production behaviour is of course conceivable (Drews, 2004), too, and must be born in mind when designing a biological production process.

4.1.2 Rheology and oxygen transfer

Sufficient oxygen supply is crucial for successful process operation in all aerobic bioprocesses. A specific level of oxygen concentration is required for directing nutrient flow towards optimal production of the required metabolite. In wastewater treatment, oxygen supply causes considerable operation costs and must be minimised. The half-saturation constant for dissolved oxygen in aerobic wastewater treatment has been reported to be in the range of 0.3 - 1.3 mg L^{-1} (Charley et al., 1980).

High biomass concentrations give rise to non-Newtonian behaviour with high apparent viscosities (see Fig. 11). This can largely be attributed to the fact that cross-linked filamentous organisms and flocs are present in the sludge. These high apparent viscosities impede oxygen transfer and the degree of mixing.



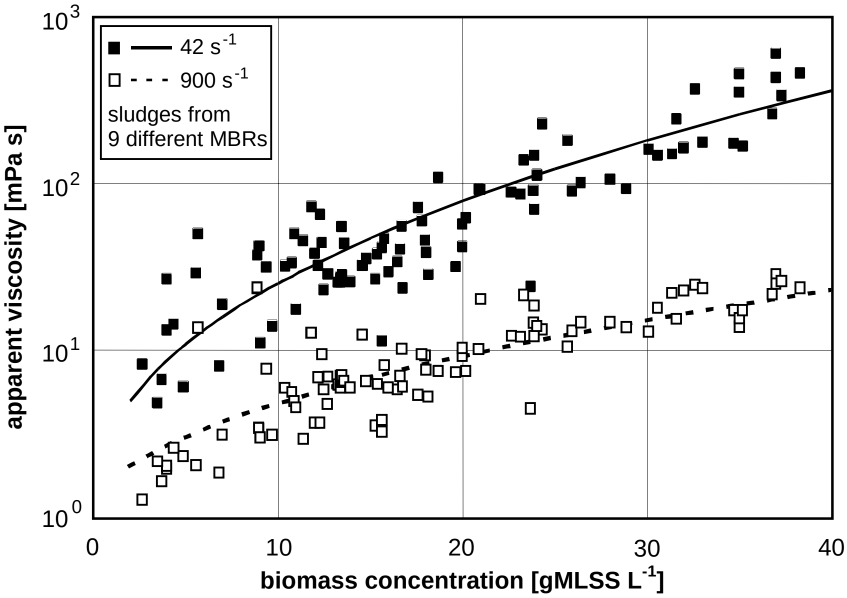


Fig. 11: Sludge structure and rheology (Rosenberger et al., 2002).

The oxygen transfer rate is described by the transfer coefficient k_La and the driving oxygen concentration difference between the surface (saturation) and the bulk of the liquid.

$$\dot{m}_{O_2} = k_L a \cdot \left(c'_{O_2} - c_{O_2}\right) \tag{5}$$

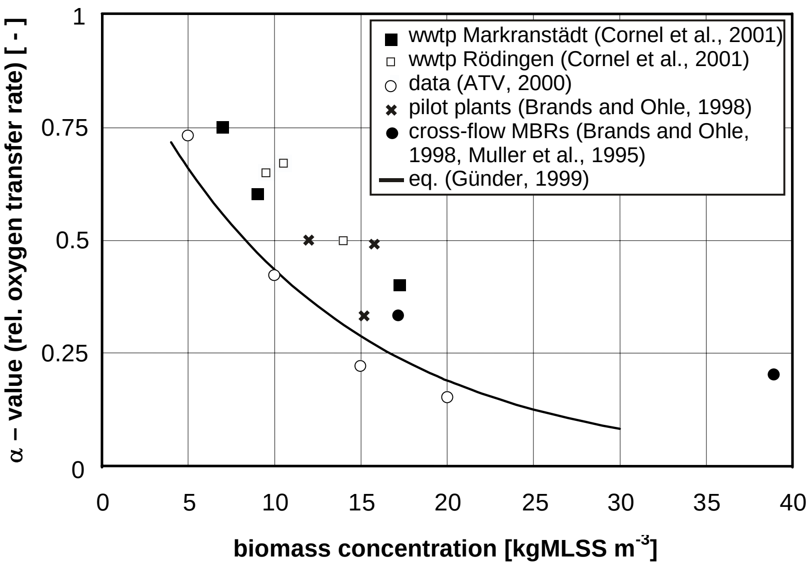


Fig. 12: Relative oxygen transfer rate (α-value) as a function of biomass concentration in MBRs

$$\alpha = \frac{k_L a_{sludge}}{k_L a_{water}} \tag{6}$$

Fig. 12 shows the impact of biomass concentration on the relative oxygen transfer rate α. Despite the different origin of data and the resulting scatter, all points show the same tendency: α, i.e. $k_L a_{sludge}$ decreases with rising biomass concentration. In order to achieve a feasible α, currently MLSS concentrations of around 15 g L^{-1} are employed for most effective MBR operation in full scale plants (Kraume and Bracklow, 2003).

4.2 Membrane operation

Membrane permeability is strongly influenced by sludge characteristics. At similar filtration conditions, Laubach (2001) found permeabilities of different sludge samples ranging from 8 to 180 L (m^2 h bar)$^{-1}$. Membrane performance can be impaired by several factors. Firstly, a cell layer forms on the membrane which can be kept small by shear stress. This, however, can become adherent as a biofilm. Besides particulate deposits, adsorption of colloidal substances causes a drop in flux. Biological factors such as nutrient levels and cell age as well as operating conditions such as shear stress due to high crossflow velocities effect the production of extracellular polymeric substances (EPS), the major cause of membrane fouling in biological applications (Rosenberger, 2003; Lesjean et al., 2004). Sludge contains different types of EPS, one fraction being dissolved or freely suspended in the liquid and the other being bound to the bacterial flocs. While Mikkelsen and Keiding (2002) state that a high concentration of bound EPS is favourable for sludge dewaterability as it causes larger flocs, Houghton et al. (2001) found an optimum bound EPS concentration for membrane filtration. To assess the filterability of a sludge, Rosenberger carried out test cell trials under defined conditions and used the filtration index I_{40} which is the flux reached in sludge filtration after 40 min divided by the clean water flux:

$$I_{40} = \frac{J_{sludge,40\,min}}{J_{water}} \tag{7}$$

As can be seen from Fig. 13, the filterability of a sludge strongly depends on the concentration of dissolved EPS.

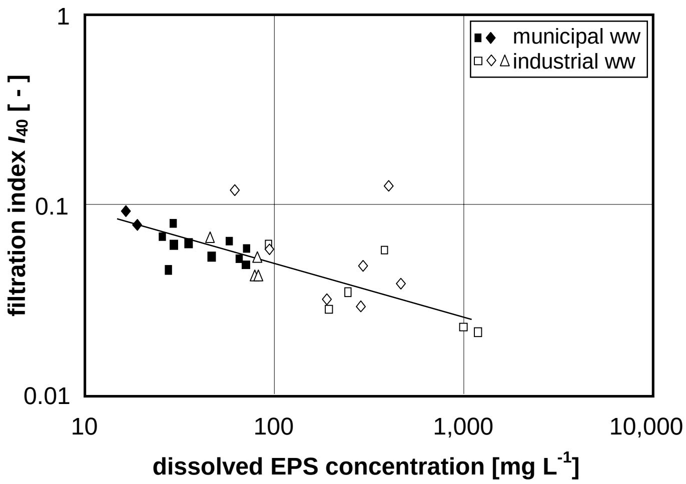


Fig. 13: Dependence of filterability of different sludges on dissolved EPS concentration (Rosenberger and Kraume, 2003; Rosenberger 2003).

The concentration of dissolved EPS depends on the type of wastewater (municipal, industrial, domestic, etc.), sludge loading rate (see Fig. 14), MLSS concentration, and mechanical stress. Due to the flow inside membrane moduls, shear in MBRs is higher than in ASP. So the EPS formation is enhanced in the case where it is least wanted, giving a closed interaction loop (see Fig. 5). Also, unsteady states like shifts in oxygen supply seem to give rise to an increased EPS formation.

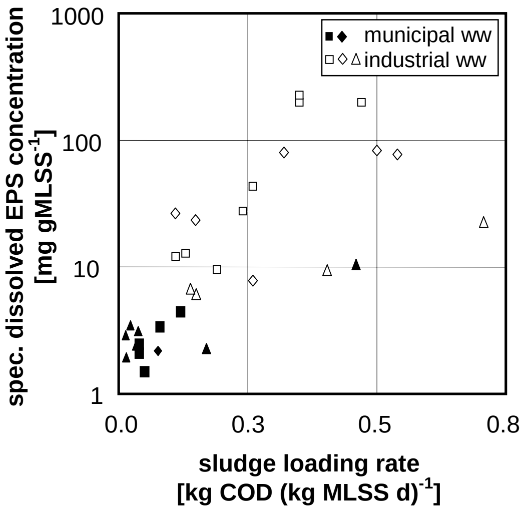


Fig. 14: Dependence of dissolved EPS concentration in different sludges on loading rate (Rosenberger, 2003).

Recents studies have revealed a linear relationship between fouling rate (increase of filtration resistance over time) and polysaccharide concentration (Lesjean et al., 2004). The authors showed that humic and low molecular weight substances pass the membrane and are therefore not responsible for fouling while polysaccharides, proteins and organic colloids are retained almost completely. This is in agreement with Chu and Li (2004) who found that the sludge cake EPS contained more polysaccharides than the suspended sludge EPS, which contained a higher fraction of TOC and proteins. Nagaoka et al. (2001) studied EPS-formation at intermittent feeding and found that the fraction of EPS formed mainly under severe substrate limitation has a higher fouling potential than the fraction formed during substrate utilisation. What mechanism causes the drop in flux – adsorption of smaller molecules and reduction of effective pore size or pore blocking by larger ones – remains unclear to date.

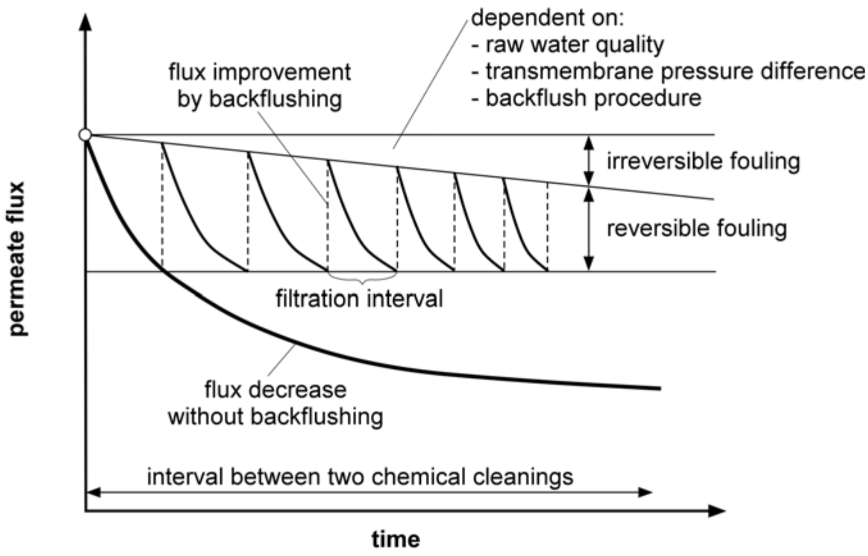


Fig. 15: Flux development

In order to remove the deposit layer, backflushing from the permeate side is employed at regular intervals during normal operation for approx. 15 – 60 s every 3 – 10 min. However, as can be seen from Fig. 15, irreversible fouling occurs over time which can only be removed by chemical cleaning. Intermediate cleaning is currently carried out every 2 – 7 days and main cleaning once or twice a year (Kraume and Bracklow, 2003).

5 Conclusions

MBRs offer new possibilities for bioprocesses and wastewater treatment according to the particular goal and demands of the process. E.g., by individual control of hydraulic and biomass residence times, production or degradation kinetics can be optimised beyond chemostat performance. Even slowly growing microorganisms with particular degradation features can be established in a mixed culture leading to better substrate and nutrient removal efficiencies. High biomass concentrations result in higher reaction rates or reduced reactor volumes. The utilization of maintenance energy demands offers the possibility of decreased excess sludge formation.

In MBRs, bioreactor and membrane filtration cannot be regarded as individual unit operations as they interact in a number of ways. Therefore, MBRs need to be considered as hybrid reactors. Existing models do not account for effects at very low growth rates typical for MBRs, so kinetic parameters determined in common experiments are not applicable anymore thereby rendering a reliable process design of MBRs on the basis of standard data impossible. No generally valid relationship for estimating maintenance energy requirements at very low growth rates in MBRs has been reported so far which highlights the need for further research. The mechanisms leading to membrane fouling are not yet known. EPS formed by biomass seems to be the major cause for fouling but neither the exact conditions that lead to its formation nor the membrane interaction are completely understood.

Although several practical experiences and data are available for design and operation of MBR processes there is still considerable optimisation potential. Some of the biology-membrane interactions are currently being investigated in detail by different groups but the overall consequences and the combination of results has yet to be done. Due to the complexity of the system, so far no coupling of biological and fouling models has been undertaken. From this, even further process improvement by application of MBRs can be expected.

Nomenclature

Symbols

c	concentration	[g L^{-1}]
I	filtration index	[-]
J	flux	[L m^{-2} h^{-1}]
k_La	transfer coefficient	[s^{-1}]
K_S	half-saturation constant	[g L^{-1}]
m	maintenance coefficient	[g L^{-1}]
$\dot{m}$	mass flux	[g $(L\ s)^{-1}$]
$\dot{r}$	reaction rate	[g $(L\ h)^{-1}$]
Y	yield coefficient	[-]
α	rel. oxygen transfer rate	[-]
μ	specific growth rate	[h^{-1}]
σ	specific substrate uptake rate	[h^{-1}]

Sub-/Superscripts

B	biomass
g	growth (true yield)
S	substrate
max	maximum
min	minimum
0	feed
’	saturation

Abbreviations

ASP	activated sludge process
ATV	Abwassertechnische Vereinigung
BOD_5	biological oxygen demand in 5 days
COD	chemical oxygen demand
EPS	extracellular polymeric substances
MBR	membrane bioreactor
MLSS	mixed liquor suspenden solids
VSS	volatile suspended solids
ww	wastewater
wwtp	wastewater treatment plant

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