Membrane Bioreactors for Wastewater Treatment

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PREFACE

Research into combining membranes with biological processes for wastewater treatment began over 30 years ago, and membrane bioreactors have been used commercially for the past 20 years. Today, over 500 membrane bioreactor processes have been commissioned to treat both industrial and municipal wastewaters, as well as for in-building treatment and re-use of greywater. In recent years the number of papers in journals, published case studies and conferences dedicated to these processes has risen exponentially. These meetings have brought together relevant biological and membrane fundamentals, the latest academic research findings, process developments and operational experiences from around the world. This text is the first to attempt to bring together all this knowledge in one book.

It is intended that this book acts as a resource for a wide range of people in the water and wastewater field, including students and researchers, consultants and engineers, operators and managers, who have an interest in membrane biorecators. If you have a background in biological processes, chapter 2 covers fundamentals of membranes required to understand membrane bioreactors. If your main expertise is in membrane systems, chapter 3 outlines key fundamentals of biological processes needed. The remainder of the book focuses on the different types of membrane bioreactor processes available: their design and operation, the latest developments and current academic understanding (Chapter 4 and 5) and their commercial application (Chapters 6 and 7).

We could not have prepared this text without the co-operation of many individuals. In particular, we thank Steve Churchouse (MBR Technology, UK), Pierre Cote (Zenon Environmental Ltd, Canada), Tony Robinson (Wehrle Werk, UK), Heinz Strohwald (EnVig Wier, South Africa) and Ludovic Huiterol (Rhodia, France) for their help in providing information for Chapters 6 and 7. Finally, we would like to thank all in Water Sciences at Cranfield University who have helped us complete the book.

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Dr Bruce Jefferson is a Research Officer in Water Sciences. He obtained his first degree in Chemical Engineering at Loughborough University, and completed his PhD there in 1996 on dissolved air flotation. He joined Cranfield as a Research Officer, and his work includes fundamentals of biomass separation membrane bioreactors and their use in water recycling.

Dr Keith Brindle has an MSc in Applied Microbiology and Biotechnology from Wolverhampton University. His PhD, at Cranfield University, investigated the application of membrane aeration bioreactors. Dr Brindle now works for the UK-based international charity WaterAid.

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Introduction

1.1 TYPES OF MEMBRANE BIOREACTOR

Combining membrane technology with biological reactors for the treatment of wastewaters has led to the development of three generic membrane bioreactors (MBRs): for separation and retention of solids; for bubble-less aeration within the bioreactor; and for extraction of priority organic pollutants from industrial wastewaters. Membranes when coupled to biological processes are most often used as a replacement for sedimentation i.e., for separation of biomass (Figure 1.1a). Such systems are well documented (Brindle and Stephenson, 1996). However, membranes can also be coupled with bioprocesses for wastewater treatment in two other ways. Firstly, they can be used for the mass transfer of gases (such systems are not to be confused with so-called 'membrane aerators' which is a term used for some fine bubble diffusers), usually oxygen for aerobic processes (Figure 1b). Secondly, membranes can used for the controlled transfer of nutrients into a bioreactor or the extraction of pollutants from wastewaters which are untreatable by conventional biological processes (Figure 1c). The target pollutants are then removed in a reactor with the correct environmental conditions for biological treatment (Brindle and Stephenson, 1996).
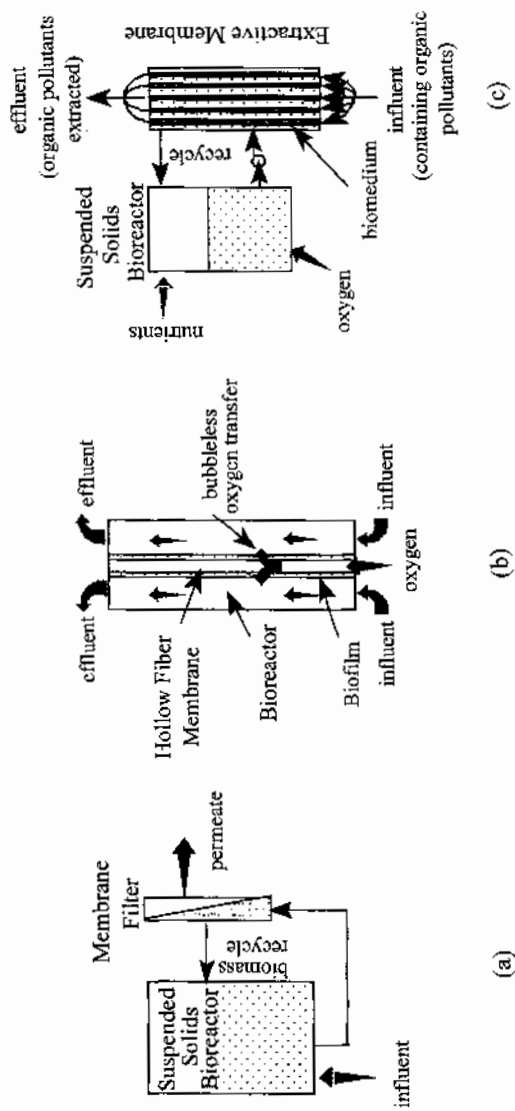
Figure 1.1. Main features of the three different MBR processes: (a) solid–liquid separation MBR; (b) oxygen mass transfer membrane bioreactor, in this case through a single hollow fibre with attached biofilm growth and (c) extractive membrane bioreactor (EMBR).

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1.2 EARLY DEVELOPMENT OF MEMBRANE BIOREACTORS FOR WASTEWATER TREATMENT

Ultrafiltration as a replacement for sedimentation in the activated sludge process was first described by Smith *et al.*, (1969). In another early report, Hardt *et al.*, (1970) used a 10 l aerobic bioreactor treating a synthetic sewage with a dead end ultrafiltration membrane for biomass separation. The mixed liquor suspended solids concentration was high compared to conventional aerobic systems at 23 to 30,000 mg l^{-1}. The membrane flux was 7.5 l m^{-2} h^{-1} and chemical oxygen demand (COD) removal was 98%. Dorr-Oliver Inc developed the Membrane Sewage Treatment (MST) process in the 1960s (Bemberis *et al.*, 1971). In the MST system, wastewater entered a suspended growth bioreactor where flow was continuously withdrawn via a rotating drum screen to an ultrafiltration membrane module. The membrane configuration was plate and frame and operated at inlet and outlet pressures of 345 kN m^{-2} and 172 kN m^{-2} respectively, achieving a flux rate of 16.9 l m^{-2} h^{-1}.

In the 1970s the technology first entered the Japanese market through a license agreement between Dorr-Oliver and Sanki Engineering Co. Ltd. By 1993, 39 of these external membrane bioreactor systems had been reported for use in sanitary and industrial applications (Aya, 1994). Today membrane bioreactor (MBR) systems used widely in Japan with several companies offering processes for domestic wastewater treatment and reuse, and some industrial applications, mainly in the food and beverage industries where high COD wastes are common.

Around the same time Thetford Systems, now part of Zenon Environmental, launched their version of an external membrane biomass separation system, the 'Cycle-Let' process, for the aerobic treatment of domestic wastewater. In the late 1980s to early 1990s Zenon Environmental continued the early development of Dorr-Oliver in developing systems for industrial wastewater treatment, resulting in two successful patent applications (Tonelli and Canning, 1993; Tonelli and Behmann, 1996). Zenon's commercial system, ZenoGem was subsequently introduced in 1982.

In 1989 the Japanese Government joined with many of the large companies to invest in the development of a low footprint, high product quality process that would be suitable for water recycling. This was in part demonstrated through the Aqua Renaissance programme '90 (Kimura, 1991). One of the participating

companies was Kubota, and they developed a flat plate immersed MBR (Churchouse and Wildgoose, 1999).

In 1982, Dorr-Oliver introduced the Membrane Anaerobic Reactor System (MARS) for the treatment of high strength food industry. The process used an external ultrafiltration membrane with an overall loading of 8 kgCOD m^{-3} d^{-1}, achieving up to 99% removal of COD. Around the same time in the early 1980s two systems were being developed in the UK with either ultra-filtration or micro-filtration membranes (Choate *et al.*, 1983; Anderson *et al.*, 1986). This concept has been further developed in South Africa in the Anaerobic Digester Ultra Filtration process (ADUF). The ADUF system employs tubular polyethersulphone membrane operating at a steady state flux of 37.3 $l.m^{-2}.d^{-1}$ and a solids concentration of 50g TSS l^{-1} (Botha *et al.*, 1992).

While biomass separation MBRs have been extensively applied at full-scale, membrane aeration bioreactors (MABRs) and extractive membrane bioreactors (EMBRs) have only been operated at up to pilot-scale. Both exploit the ability of a membrane to separate two distinct phases, while allowing transport of components from one phase to the other, to optimise conditions within the bioreactor for enhanced microbial degradation of wastewater pollutants. One of the earliest reports of a bubble-less gas transfer was for oxygenation of wastewaters using dense membranes (Yeh and Jenkins, 1978). The EMBR concept has been developed to treat chlorinated organic compounds present in aggressive waste streams with low pH and high total dissolved solids (Livingston, 1994).

1.3 THE CURRENT STATUS OF MEMBRANE BIOREACTORS FOR WASTEWATER TREATMENT

Full-scale commercial aerobic MBR processes first appeared in North America in the late 1970s and then in Japan in the early 1980s, with anaerobic processes entering the industrial wastewater market at around the same time in South Africa. The introduction of aerobic MBRs into Europe did not occur until the mid-1990s.

There are over 500 commercial MBRs in operation worldwide, with many more proposed or currently under construction. Commercial MBRs have proliferated in Japan, which has approximately 66% of the world's processes (Table 1.1). The rest are predominately either in North America or Europe. Over 98% of these systems couple the membrane separation process to an aerobic biological process rather than to an anaerobic bioreactor. Approximately 55% of

these commercial systems have the membrane submerged within the bioreactor while the remainder have the membrane units external to the biological process.

Table 1.1: Approximate global distribution of MBRs by wastewater type. Number of plants shown as a percentage of the total number of membrane processes.

Type of wastewater	Approximate % of total MBRs
Industrial	27
In-building	24
Domestic	27
Municipal	12
Landfill leachate	9

Of the six commercial MBRs described in this book, five are aerobic and one anaerobic, four have the membrane units submerged in the bioreactor while the two remaining have their membrane units external to the bioreactor (Table 1.2). The MBRs that have sidestream membranes tend to operate at higher crossflow velocities, transmembrane pressures (TMPs) and permeate flux rates compared to the submerged membrane units. Though the manufacturers of such units argue that sidestream membrane designs are much easier to retrofit to existing units, the membranes usually require a more demanding cleaning protocol.

The different commercial MBR processes achieve the TMP necessary for permeation by a number of different ways. The Kubota MBR uses hydrostatic head above the membrane unit, which is submerged at depth, during normal operation. A combination of hydrostatic head and a vacuum applied to the permeate side of the membrane is used in the ZenoGem MBR and the Kubota process during peak hydraulic loading. Finally, throttling the pressure in the recirculation line to which the sidestream membrane unit is incorporated is used by Orelis and Wehrle Werk MBR processes.

Table 1.2: Summary of commercial MBRs

	Bioreactor	Type	Membrane	Flux ($1\ m^{-2}\ h^{-1}$)
Kubota	aerobic	submerged	flat panels	25
Zenon	aerobic	submerged	hollow fibre	30
Orelis	aerobic	sidestream	flat panels	100
USF	aerobic	submerged	tubular	40
Membratek	anaerobic	sidestream	tubular	40
WehrleWerk	aerobic	sidestream	tubular	100

The aerobic MBR process has successfully treated effluents from a range of industrial wastewaters, including cosmetics, pharmaceuticals, metal fabrication, textiles, abattoirs, dairy, food, beverage, paper and pulp, rendering and chemical manufacture. Landfill leachate treatment by the aerobic MBR has also been undertaken in Europe.

The very high quality of the treated water from an MBR process is common to all commercial aerobic systems. Complete solids removal, a significant disinfection capability, high rate and high efficiency organic and nutrient removal and a small footprint are all characteristics of the MBR, regardless of the wastewater type to be treated or the commercial process used. These universal attributes are due to the membrane and biological synergism described in Chapter 4. The quality of the treated water from these processes is so high that recycling and reuse is often a viable option. The wastewater treated to reuse standards includes blackwater as well as greywater. From large shopping malls in New Jersey, USA, to the YMCA in Tokyo, Japan, commercial MBRs are providing a high performance, low maintenance, small footprint on-site process capable of a high quality treated water suitable for reuse (Chapter 6). More recently a reduction in the cost of the membrane modules combined with extended life expectancy of the membranes, acceptance of the processes capabilities, advances in process design and operation have all resulted in the MBR process treating large volume, low strength municipal wastewaters.

Neither the EMBR nor the MABR have yet to be commercially exploited. Research and development of both processes centres around a few universities and spin-out companies in the USA and Europe.

1.4 ADVANTAGES OF MEMBRANE BIOREACTORS

Table 1.3 below lists the principle advantages and disadvantages of the different MBRs. These advantages and disadvantages are considered in detail in the remainder of this book. However, it is clear that the combining of membrane process with biological wastewater treatment processes resulted in advantages that were once exclusive to the former; particularly small footprint, process intensification, modular, and retrofit potential.

Table 1.3: Advantages and disadvantages of MBRs

Advantages	Disadvantages
Membrane Separation Bioreactors	
Small footprint	Aeration limitations
Complete solids removal from effluent	Membrane fouling
Effluent disinfection	Membrane costs
Combined COD, solids and nutrient removal in a single unit	
High loading rate capability	
Low/zero sludge production	
Rapid start up	
Sludge bulking not a problem	
Modular/retrofit	
Membrane Aeration Bioreactors	
High oxygen utilisation	Susceptible to membrane fouling
Highly efficient energy utilisation	High capital cost
Small footprint	Unproven at full-scale
Feed-forward control of O demand	Process complexity
Modular/retrofit	
Extractive Membrane Bioreactors	
Treatment of toxic industrial effluents	High capital cost
Small effluents	Unproven at full-scale
Modular/retrofit	Process complexity
Isolation of bacteria from wastewater	

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2

Membrane fundamentals

2.1 THE MEMBRANE

2.1.1 Membrane definition

A membrane can be thought of as a material through which one type of substance can pass more readily than others, thus presenting the basis of a separation process. It is thus the property of the membrane to separate components of the water to be treated that is of key interest when selecting or designing membrane separation systems for such duties arising in the water industry. For many processes the membrane acts to reject the pollutants, which may be suspended or dissolved, and allow the 'purified' water through it. In some cases, however, the membrane may act so as to extract pollutants from the wastewater, as in the extractive MBR (Livingston, 1994), or transfer gas to the wastewater, as in the bubble-less oxygenation MBR (Brindle *et al.*, 1998).

2.1.2 Membrane structure and categorisation

The principal objective in membrane manufacture is to produce a material of reasonable mechanical strength, and which can maintain a high throughput of a desired permeate with a high degree of selectivity. These last two parameters are mutually counteractive, since a high degree of selectivity is normally only achievable using a membrane having small pores and thus an inherently high hydraulic resistance (or low permeability). The permeability increases with increasing density of pores, implying that a high material porosity is desirable. The overall membrane resistance is directly proportional to its thickness. Finally, selectivity will be compromised by a broad pore size distribution. It stands to reason, therefore, that the optimum physical structure for any membrane material is based on a thin layer of material with a narrow range of pore size, and a high surface porosity.

The range of available membrane materials is very diverse. They vary widely both in chemical composition and physical structure, but the most fundamentally important property is the mechanism by which separation is actually achieved. On this basis membranes may be categorised as either dense or porous (Table 2.1). Separation by dense membranes relies to some extent on physico-chemical interactions between the permeating components and the membrane material, and relate to separation processes having the highest selectivity (Figure 2.1). Reverse osmosis, electrodialysis and nanofiltration processes are thus able to separate ions from water, electrodialysis being unusual in achieving membrane permeation through the application of an electromotive force, rather than transmembrane pressure. Porous membranes, on the other hand, achieve separation mechanically (i.e. ostensibly by sieving) and are thus mechanistically closer to conventional filtration processes. Ultrafiltration can remove colloidal and dissolved macromolecular species, and as such their ability to reject material is defined by the molecular weight cut-off (MWCO) in Daltons (i.e. the relative molecular weight) of the rejected solute, rather than its physical size. Microfiltration, on the other hand, is capable of removing only suspended materials—generally down to around 0.05μm in size. It is the porous membranes that are used in MBRs to retain the suspended solids material, mainly biomass, within the reactor while producing a clarified effluent (Chapter 4).
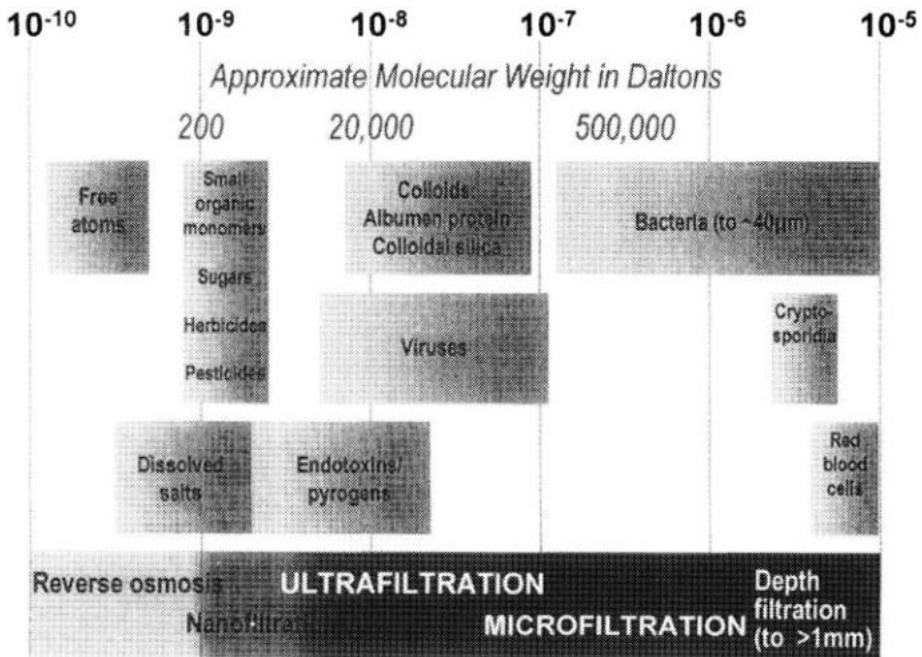
Figure 2.1. Rejection capability of the different membrane separation processes.

Table 2.1: Dense and porous membranes for water treatment

Dense	Porous
Membrane separation processes	
Reverse Osmosis (RO), or hyperfilration	Ultrafiltration (UF)
Separation achieved by virtue of differing solubility and diffusion rates of water (solvent) and dissolved species (solutes) in water (as used in EMBR processes)	Separation of both large, dissolved solute molecules and suspended colloidal particles by size exclusion.
Electrodialysis (ED)	Microfiltration (MF)
Separation achieved by virtue of differing ionic size, charge and charge density of solute ions, using ion-exchange membranes.	Separation of suspended solids from water by size exclusion (predominately used in MBRs for solid separation and MABRs for aeration)
Nanofiltration (NF)	
Formerly called leaky reverse osmosis. Separation achieved through combination of charge rejection, solubility-diffusion and sieving	
Membrane materials	
Limited to polymeric materials	Both polymeric and inorganic materials available

A more convenient practical categorisation of membranes is according to the material composition, which is generally either organic (polymeric) or inorganic (ceramic or metallic). The physical structure of the membrane based on these materials can then vary according the exact nature of the material and/or the way in which it is processed. In general, membrane materials employed in pressure-driven processes tend to be anisotropic; they have symmetry in a single direction such that their pore size varies with membrane depth. It is only the very top layer of the membrane which actually demonstrates substantial permselectivity, the remainder merely providing mechanical support.

Examples of membrane materials are listed in Tables 2.2 and 2.3. Membrane manufacture primarily concerns the production of a porous material. The cost of the membrane is therefore dependent not only the raw material but also on the ease with which pores of the desired size or size distribution can be introduced. This can vary considerably between materials and according to the precision of the pore size distribution (or degree of isoporosity).

Inorganic membranes, for example, are formed by the pressing and sintering of fine powders onto a pre-prepared porous support. This tends to be a very expensive process, particularly if a membrane layer of even thickness and narrow pore size distribution is to be produced. The cost of microfiltration or ultrafiltration membranes derived from titanium and/or zirconium may exceed £1000 per m^2. At the opposite end of the spectrum are simple, homogenous

polymeric membranes produced by extrusion (stretching) of partly crystalline sheets perpendicular to the orientation of crystallites, possibly with the assistance of a fibrulating agent, such as microscopic glass beads, to promote the formation of pores. These microporous materials cost less than £10 per m^2 to produce, but are limited in their permeability, isoporosity and mechanical strength.

Many polymeric microporous membranes are produced by phase inversion. A polymeric solution is cast to produce a thin layer of material. The porous side of the membrane is produced by precipitation of the polymer in water, a process sometimes referred to as gelation. The permselective side of the membrane is produced by evaporation of the polymer solvent to produce a 'skin' of much lower permeability, resulting in an anisotropic membrane structure.

Table 2.2: Membrane materials by type

Membrane	Manufacturing procedure	Structure	Applications
Ceramic	Pressing, sintering of fine powders	0.1–10 µm pores	MF, gas separation, separation of isotopes
Stretched	Stretching of partly crystalline foil	0.1–1 µm pores	Filtering of aggressive media, sterile filtration, medical technology
Etched polymers	Radiation followed by acid etching	0.5–10 µm cylindrical pores	Analytical and medical chemistry, sterile filtration
Supported liquid	Formation of liquid film in inert polymer matrix	Liquid-filled porous matrix	Gas separations, carrier-mediated transport
Symmetric microporous	Phase inversion reaction	0.05-5 µm pores	Sterile filtration, dialysis, membrane distillation
Integral asymmetric microporous	Phase inversion reaction followed by evaporation	1–10 nm pores at membrane surface	UF, NF, gas separation, pervaporation
Composite asymmetric microporous	Application of thin film to microporous membrane	1-5 nm pores at membrane surface	UF, NF, gas separation, pervaporation
Ion exchange	Functionalisation of polymer material	Matrix of positive and negative charges	ED

Table 2.3: Membrane materials by name

Polymer	Advantage	Disadvantages
Titanium dioxide/ Zirconium dioxide	Good thermal resistance Good chemical resistance Good mechanical resistance	Very expensive Limited to MF and UF Brittle materials
Cellulose acetate	Inexpensive Chlorine resistant Solvent cast	Poor thermal stability Poor chemical stability Poor mechanical stability
Polysulphone	Steam sterilisable pH resistant Solvent cast	Poor resistance to hydrocarbons
Polypropylene	Chemically resistant	Hydrophobic unless surface treated
PTFE	Very hydrophobic Excellent organic resistance Excellent chemical stability Sterilisable	Very hydrophobic Expensive
Polyamide	Good chemical stability Good thermal stability	Sensitive to chlorine

2.1.3 Membrane configurations

The geometry of the membrane, i.e. the way it is shaped, is crucial in determining the overall process performance. Other practical considerations concern the way in which the individual membrane elements, that is the membranes themselves, are housed to produce modules. The optimum geometry, or configuration, for an individual membrane element is one that has the following characteristics:

- a high membrane area to module bulk volume ratio;
- a high degree of turbulence for mass transfer promotion on the feed side;
- a low energy expenditure per unit product water volume;
- a low cost per unit membrane area;
- a design that facilitates cleaning;
- a design that permits modularisation.

Some of these characteristics are mutually exclusive. For example, promoting turbulence results in an increase in the energy expenditure. Furthermore, direct mechanical cleaning of the membrane is only possible on comparatively low area: volume units where the membrane is accessible. It is not possible to

produce a high-membrane area to module bulk volume ratio without producing a unit having narrow feed channels, which will then adversely affect the cleaning regime and turbulence promotion.

There are five principal configurations currently employed in membrane processes which all have various practical benefits and limitations (Table 4.2). The configurations are based on either a planar or cylindrical geometry and comprise:

- pleated filter cartridge;
- plate-and-frame;
- spiral-wound;
- tubular;
- hollow fine fibre.

Of the flat plate geometries the lowest cost is the pleated filter cartridge, used exclusively in microfiltration and generally designed as a disposable unit. The most commercially significant application of the plate and frame design is in electrodialysis modules (or stacks), although some microfiltration units and one reverse osmosis module design are also based on this configuration. Spirally-wound membranes have the advantage of a simple construction while providing a reasonable membrane area per unit bulk, and is the standard configuration for reverse osmosis and nanofiltration modules.

Wide-bore tubular membranes (in excess of ~5 mm diameter) are preferred when turbulence promotion is important (see Section 2.2.2) and/or when frequent mechanical cleaning is necessary. Most inorganic membranes are based on this geometry, often in the form of monoliths: cylindrical channels in solid ceramic matrices. For polymeric materials the inherent radial mechanical strength of the tube increases with decreasing diameter such that at a certain diameter, which depends upon the wall thickness, the tube becomes self-supporting for both out-to-in and in-to-out flow and thus can be backflushed (i.e. cleaned by flow reversal). Hollow fibres are self-supporting membranes providing the highest membrane area per unit volume at the lowest cost, and are usually operated out-to-in with reference to water permeation (but can operate in-to-out for aeration), and are periodically backflushed. Such backflushing can demand relatively extreme conditions, such as the use of compressed air.

2.1.4 MBR membrane materials and configurations

The development or selection of membrane materials and module configurations for MBR applications has been governed by the need to suppress membrane fouling (Section 2.2.4.2) or ameliorate the problems associated with it. The nature and extent of fouling depend both upon the feedwater quality and specific facets of the membrane separation system. For a conventional pressure-

driven process the latter comprise mainly the membrane material itself, the membrane element representative linear dimension (i.e. the channel height or the tube diameter), the specific permeation rate (or flux) and the hydrodynamic conditions prevailing at the membrane solution interface.

As with most other membrane applications, the preferred membrane materials for MBRs are invariably polymeric on the simple basis of cost (Table 2.5). Geometries employed in key commercial systems range from flat plate/plate and frame (Kubota, Japan, Rhodia Pleiade-based MBR system, France) to tubular (Milleniumpore, UK) or hollow fibre (Zenon, Canada). The choice of configuration is profoundly influenced by the MBR process configuration, namely by whether the membrane element is placed within the bioreactor or external to it (Chapter 4). Gas transfer and extractive systems, on the other hand, rely on maximising the membrane surface area, such that hollow fibre is the configuration of choice for these systems (Chapter 5).

Table 2.4: Membrane configurations

Configuration	Area/volume Ratio (m^2/m^3)	Cost	Turbulence Promotion	Advantages	Disadvantages	Applications (most important first)
Pleated cartridge	800–1000	Low	Very poor	robust construction compact design	easily fouled cannot be cleaned	Dead end MF
Plate-and-frame	400–600	High	Fair	can be dismantled for cleaning	complicated design cannot be backflushed	ED, UF, RO
Spiral-wound	800–1000	Low	Poor	low energy cost robust and compact	not easily cleaned—cannot backflush	RO, UF
Tubular	20–30	Very high	Very good	easily mechanically cleaned tolerates high TSS waters	high capital and membrane replacement cost	Cross-flow filtration, high TSS waters
Capillary tube	600–1200	Low	Good	characteristics between tubular and hollow fibre		UF
Hollow fibre	5000–40000	Very low	Very poor	can be backflushed compact design tolerates high colloid levels	sensitive to pressure shocks	MF, RO

NB   The capillary tube is used in UF, where the water flows from inside to outside the tubes.
        The hollow fibre is used in RO and MF, where the water flows from outside to inside the tubes.24

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Table 2.5: Membranes and configurations for MBR

MBR Type	Membrane Material	Configuration	Pore Size (μm)	Reference
Solid Separation	polyethylene	Hollow fibre	0.4	Ueda and Hata, 1991
	ceramic	Tubular	0.1	Ghyoot et al., 1999
	polysulphone	Tubular	0.1	Ghyoot et al., 1999
	polyethylene	Plate and frame	0.4	Jefferson et al., 2000
	polypropylene	Plate and frame	5	Gander et al., 2000
	polyethylene	Hollow fibre	0.2	Wilkes et al., 2000
Aeration	polytetrafluoroethylene	Hollow fibre	2	Suzuki et al., 1993
	polypropylene	Hollow fibre	0.04–1.0	Pankhania et al., 1994
	polytetrafluoroethylene	Plate and frame	0.2	Timberlake et al., 1988
	Silicone	Hollow fibre	Dense	Hirasa et al., 1991
Extraction	Polysulphone & ZnO_2		UF	Diels et al., 1993 Peys et al., 1997
	silicone	Tubular	Dense	Brookes and Livingston, 1993

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2.2 THE PROCESS FUNDAMENTALS

There are essentially two aspects to membrane systems behaviour: steady state and dynamic. Most membrane processes are configured so as to encourage the attainment of steady state conditions, although some are designed for batch operation with a regeneration cycle. However, it is very common for systems which, in principle, would be expected to reach steady-state to undergo a gradual deterioration in performance. The reasons for this, along with definitions of the basic process performance parameters, are given below.

2.2.1 Process definitions

2.2.1.1 Flux and crossflow

The key elements of any membrane process are the influence of the following parameters on the overall permeate flux:
- the membrane resistance;
- the operational driving force per unit membrane area;
- the hydrodynamic conditions at the membrane:liquid interface;
- the fouling and subsequent cleaning of the membrane surface.

The flux is the quantity of material passing through a unit area of membrane per unit time. This means that it takes SI units of m^3 per m^2 per s, or simply metres per second, and as such is sometimes referred to as the permeate velocity. Other non-SI units used are litres per m^2 per hour (or $1\ m^{-2}\ h^{-1}$) and m per day. The flux is determined by both the driving force and the total resistance offered by the membrane and the interfacial region adjacent to it. The resistance of the membrane is fixed, unless it becomes partly clogged (or fouled internally) by components in the feed water. The interfacial region resistance is, on the other hand, a function of both feedwater composition and permeate flux since, for a conventional pressure driven process, the materials rejected by the membrane tend to accumulate within the interfacial region at a rate dependent on the flux (Section 2.2.4). These materials may then foul the membrane through a number of physicochemical mechanisms. The process operational efficiency is therefore determined by the extent to which the forces opposing the driving force predominate.

In most membrane processes there are three streams: a feed, a retentate and a permeate stream. The retentate stream is unpermeated product. If there is no retentate stream then operation is termed dead-end or full-flow (Figure 2.2). Such operation is normally restricted to either low-solids water, such as

cartridge filtration of boiler feedwater or ultrafiltration for apyrogenic water production, or cyclic operation with frequent backwashing, such as the Memcor microfiltration process. The alternative to dead-end operation is crossflow operation (Figure 2.2), in which the feedwater flows parallel to the membrane surface and so expediting the removal of accumulated materials from the membrane: solution interfacial region. Crossflow operation then implies the existence of a retentate stream. The more perm-selective the membrane, and hence the larger the hydraulic resistance, the greater the propensity for crossflow rather than dead end operation. Thus, while MF and UF membrane elements may be dead-ended under the appropriate conditions, RO and NF membranes may not.

2.2.1.2 Conversion

The combination of the flux and the total membrane area determine the *conversion* or *recovery* of the process. The conversion – normally expressed as a percentage Θ – is the amount of the feed which is recovered as permeate.

For concentration C and flow Q in feed, retentate and permeate (Figure 4), a simple mass balance dictates that:

$$Q = Q_P + Q_R \qquad (4.1)$$

$$QC = Q_P C_P + Q_R C_R \qquad (4.2)$$

where % recovery or conversion is given by:

$$\Theta = \frac{Q_P}{Q} \times 100 \qquad (4.3)$$

and the subscripts P and R respectively refer to permeate and retentate.

In MBRs the permeate concentration C_P is normally very small compared to the feed level C, both with respect to suspended and oxygen demanding materials. Moreover, the sludge volume production rate Q_R is normally very small compared to the feed flow rate Q. The MBR process thus represents a very substantial elimination of biodegradable matter.
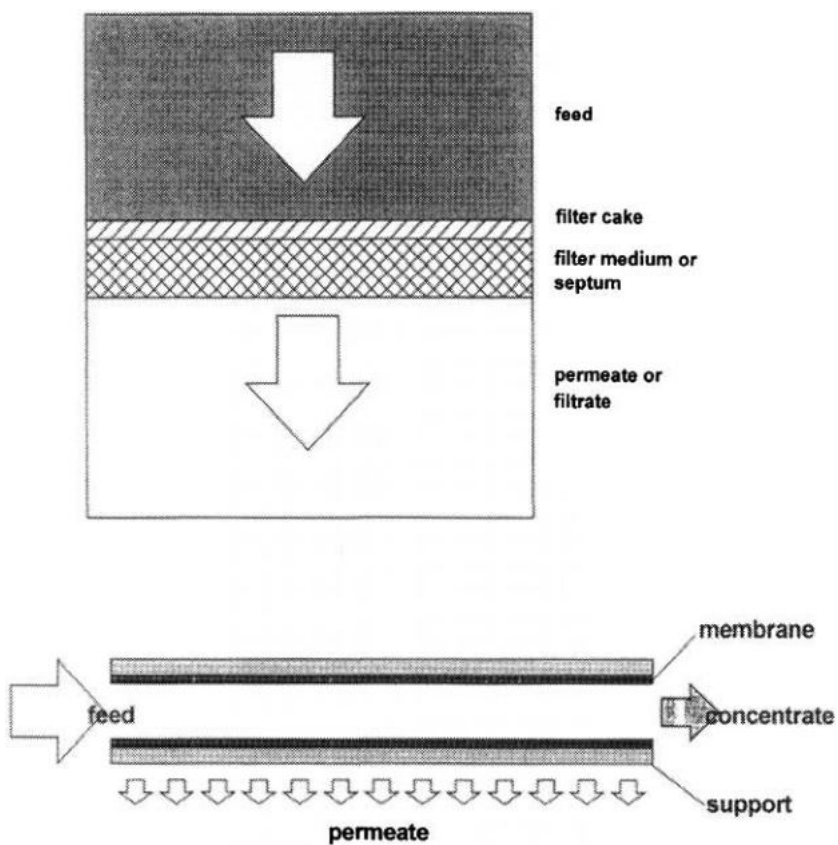
Figure 2.2. Dead-end (top) and crossflow microfiltration (bottom).
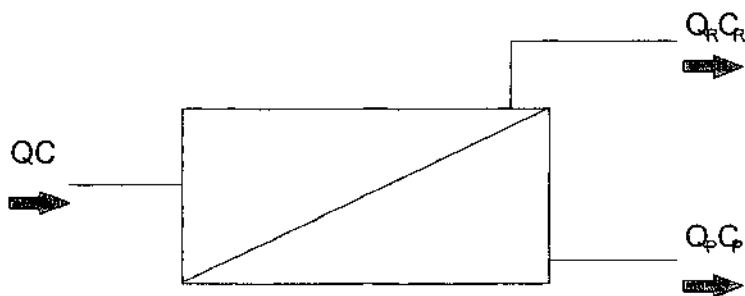
Figure 2.3. Membrane module mass balance.

2.2.1.3 Rejection

The membrane is a selective barrier: it lets some types of matter pass through it while rejecting others. This property is normally expressed as the rejection where:

$$R = \frac{1 - C_P}{C} \times 100 \qquad (4.4)$$

Note that it is possible to have negative rejection values if the membrane is selective for specific contaminants, as would be the case for an extractive membrane system. However, as stated, in a conventional perm-selective MBR rejection of suspended materials and oxygen demand is normally very high.

2.2.2 Mass transport

Permeation through a membrane demands that some force be applied to drive matter across it. The force can be natural or artificially imposed, and the two most important transport mechanisms in membrane processes for wastewater treatment are convection and diffusion.

Convection results from the movement of the bulk fluid, rather than any components dissolved or suspended in it. Hence, any flowing liquid constitutes convective transport. The type of flow produced, or the flow regime, is dependent upon the flow rate. At high flow rates the flow is described as turbulent, whereas at low flow rates it is defined as laminar. Higher flow rates usually yield greater mass transport, and hence it is always desirable to promote turbulence on the retentate side of a membrane.

Brownian diffusion results from the transport of individual ions, atoms or molecules by thermal motion. The basic law defining diffusive transport (originally developed by Fick and known as Fick's first law of diffusion) dictates that its rate is dependent upon the concentration gradient coupled with the component Brownian diffusivity, which increases with decreasing size.

2.2.3 The driving force

The driving force for a process is usually a pressure gradient, although both extractive and gas transfer systems operate via a concentration gradient. In almost all pressure-driven membrane processes applied to water treatment the desired permeate is water, such that the retained or rejected material (the retentate) is concentrated. In extractive operations the permeate is the dissolved solute and the retentate the product water. Extractive systems rely on depletion of the permeating component on the permeate side of the membrane to generate a great enough concentration gradient across the membrane to transfer matter through it via diffusion. In gas transfer processes the concentration gradient is achieved by increasing the partial pressure on the unpermeated side.

Since the flux and driving force are interrelated, either one can be fixed for design purposes. It is usual to fix the value of the flux and then determine the appropriate value for the transmembrane pressure for pressure-driven processes (Table 2.6).

Table 2.6: Driving forces applicable to membrane processes

Membrane separation process	Driving force for mass transport	Type of membrane employed (pore size)	Contaminant size	Separation achieved (permeate first)
Microfiltration (MF)	Hydrostatic pressure difference, 20–200 kPa	Isotropic or anisotropic (0.1–2μm)	0.2–100μm	Water from suspended solids (as used in MBR processes for biomass retention)
Ultrafiltration (UF)	Hydrostatic pressure difference, 50–1000 kPa	Anisotropic porous (2–50nm)	5–500nm	Water from dissolved solids or colloidal macromolecules
Reverse osmosis (RO)	Hydrostatic pressure difference, 600–10^4 kPa	Composite; homogeneous Ultrathin layer	0.2–10nm	Water from low-molecular weight components and ions
Extraction/ Aeration	Concentration difference/ pressure	Composite; homogeneous Ultrathin layer	0.2–10nm	Volatile species from water/gas into water (as used in the membrane aeration bioreactor and extractive MBR)
Dialysis	Concentration difference	Homogeneous	50–5000nm	Low-molecular weight components from macromolecules
Electro-dialysis (ED)	Difference in electrical potential	Ion exchange membrane	<0.1–0.5nm	Ions from water

2.2.4 Factors opposing the driving force

The overall resistance of the membrane and interface is increased by a number of factors, each of which places an important constraint on the design and operation of membrane process plant:

- the concentration of rejected solute (as in RO and UF) or permeated ions (as in ED) near the membrane surface;
- the depletion of ions near the membrane surface (as with ED);
- the precipitation of, normally, macromolecular species at the membrane surface (gel layer formation);
- the accumulation of retained solids on the membrane (as in MF); and
- the accumulation of foulants on or within the membrane.

2.2.4.1 Concentration polarisation

Concentration polarisation (CP) is the term given to describe the tendency of the solute to accumulate at membrane:solution interface within a concentration boundary layer or (stagnant) liquid film (Figure 2.4). This layer contains near stagnant liquid, since at the membrane surface itself the liquid velocity must be zero. This implies that the only mode of transport within this layer is diffusion, which is considerably slower than convective transport in the bulk liquid region. Rejected materials thus build up in the region adjacent to membrane, increasing their concentration over the bulk value. For Brownian diffusion this build up occurs exponentially with increasing flux (Section 2.3.2). The thickness of the boundary layer, on the other hand, is determined entirely by the system hydrodynamics, decreasing in thickness when turbulence is promoted.

For pressure-driven processes operating under crossflow conditions, the greater the flux, the greater the build up of solute at interface; the greater the solute build up, the higher the concentration gradient; the steeper the concentration gradient, the faster the diffusion. These mass transfer rates are all in dynamic equilibrium with one another. CP increases the propensity for sparingly-soluble solutes to precipitate out onto the membrane, as well as generally increasing the concentration of colloidal or suspended material at the membrane surface. Furthermore, CP can increase the permeation of the rejected materials through the membrane because of the increase in the trans-membrane concentration gradient generated. It is thus always desirable to suppress CP either by promoting turbulence or running at lower fluxes.
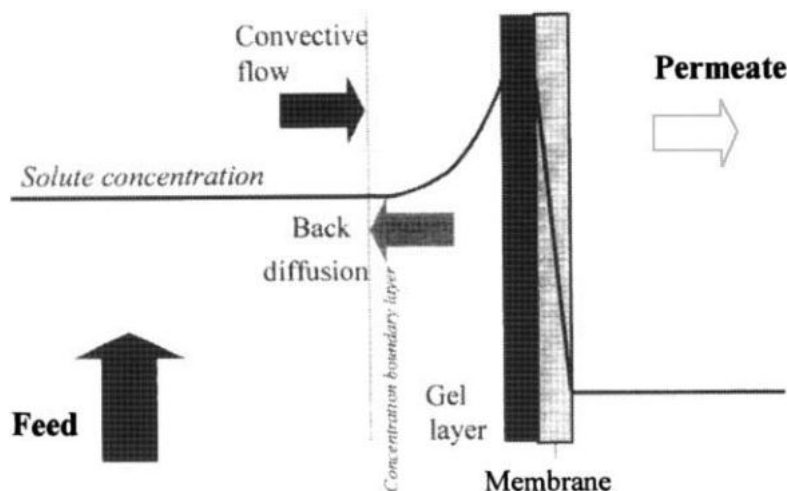
Figure 2.4. Concentration polarisation

Precipitation at the membrane:solution interface of organic solutes rejected by a UF membrane leads to the formation of a *gel layer*, which may have greater selectivity and lower permeability than the membrane itself. In such cases, the gel layer is sometimes referred to as a dynamic membrane. CP in MF processes, which employ membranes with pores capable of rejecting only suspended matter, none-the-less leads to the accumulation of suspended matter at the membrane surface to form a dynamic layer. In both cases the dynamic layer is of nominally near-constant thickness under steady state conditions, although in practice its overall hydraulic resistance is invariably time-dependent (see Section 2.2.4.2). This is to be distinguished from dead-end operation, in which the rejected solids form a filter cake which normally offers a higher hydraulic resistance than the membrane itself and which increases in thickness with time: there is no dynamic equilibrium set up since there is no retentate stream.

The relationship between driving force and polarisation in pressure-driven membrane separation processes can therefore be summarised as follows: the flow through a given type of membrane varies as the membrane area and the net applied driving force, and the power consumption is proportional to the driving force, and inversely proportional to the membrane area installed. This is analogous to electrical conduction, where the current varies with the cross-section area of copper in the cable and with the applied voltage, and the power loss in the cable varies with the voltage loss and inversely with the area. In

addition the selective nature of the process means that rejected material remains on the membrane surface. Crossflow operation affords some limitation to the extent to which rejected material accumulates in the interfacial region. These two factors are, of course, interlinked: a high driving force yields high flux and a high rate of rejected material collecting on the membrane surface, which then needs to be dispersed rapidly if the process is not to grind to a halt.

2.2.4.2 Fouling

Fouling is the general term given to the process by which a variety of species present in the water increase the membrane resistance, by adsorbing or depositing onto its surface, adsorption onto the pore surfaces within the bulk membrane material (pore restriction) or by complete pore-blocking. Mathematical models developed to represent these three mechanisms are given later (Section 2.3.2.4). Fouling can occur through a number of physicochemical and biological mechanisms, and is exacerbated by concentration polarisation since this effectively increases the concentration of foulants in the vicinity of the membrane. Fouling by individual components tends to be specific to the membrane material and application, but in general physico-chemical fouling, i.e. fouling unrelated to biological growth, can be attributed to two key components in the feed: proteins and colloidal/particulate materials.

Much research work has been carried out on membrane separation of protein because of its industrial importance, and a number of review articles are available (Fane, 1986; Howell and Nyström, 1993; Marshall *et al.*, 1993; Belfort *et al.*, 1994). Proteins can cause severe fouling of a many different MF membrane materials, in particular hydrophobic polymers such as polypropylene, causing flux declines of an order of magnitude or more (Palacek and Zydney, 1994; Judd and Till, 2000) ultimately due to irreversible deposition onto and penetration into the bulk membrane material. UF membranes, on the other hand, are somewhat less prone to fouling by macromolecules because the smaller pores are more impenetrable. For both UF and MF surface chemistry, specifically hydrophilicity and surface charge, plays an important part in determining the extent of fouling: hydrophobic materials are more prone to protein deposition leading to irreversible fouling.

Proteins can arise either as dissolved or suspended matter, but their behaviour within membrane separation systems is complex. Membrane fouling takes place both by adsorption and by deposition, with deposition being the most deleterious to performance with regards to flux decline in MF systems. It is thought that the high shear conditions prevailing at the membrane:solution interface promote aggregation of the protein or else produce other conformational changes which produce deposits of widely varying structures

and hydraulic behaviour (Kim *et al.*, 1992; Meuller and Davis, 1996). It has been proposed by Kelly *et al.* (1993) that deposited protein aggregates may serve as nucleation sites for non-aggregated, dissolved proteins. The relative extent of internal to external fouling appears to be governed as much by physical phenomena such are surface porosity and flux as by membrane surface chemistry. Meuller and Davis (1996) found high surface porosities to be deleterious to maintaining a high flux in their studies of membrane fouling of different membrane materials of the same nominal pore size. Marshall *et al.* (1997) found internal fouling to be promoted at lower fluxes.

Colloidal and particulate materials are similarly affected by physical changes in structure as a result of permeation. In this case, it is the aggregation rate which is affected by the physicochemical conditions via hydrodynamic and surface force effects. Aggregation (or flocculation) may be promoted orthokinetically (i.e. by virtue of a velocity gradient) at the membrane solution interface or within the membrane pore. Flocculation, however, is dependent upon the extent of particle destabilisation, or coagulation, generally resulting from the reduction in the magnitude of surface charge—which itself is dependent on both the bulk material properties and solution chemistry. The abstruse nature of colloidal behaviour and its sensitivity to surface forces makes particle aggregation kinetics difficult to predict, even in ideal systems. However, in their study of municipal wastewater treatment, Pouet and Grasmick (1995) identified the supracolloidal fraction, i.e. above 1 μm in particle size, as being principally responsible for fouling of a sidestream ceramic MF of sub-micron pore size.

With specific regard to filtration of activated sludge in aerobic MBRs, it is widely recognised that the main foulants are the extracellular polymeric substances (EPS) excreted from cells (Stec and Field, 1995; Chang and Lee, 1998; Nagaoka *et al.*, 1998, 1999). Chang and Lee (1998) experimentally determined that a 40% reduction in EPS (by cultivating the activated sludge under nitrogen-deficient conditions) resulted in an equivalent reduction in the hydraulic resistance of the cake. Nagaoka *et al.* (1999) similarly linked hydraulic resistance to EPS levels, including empirical parameters for EPS production and degradation in their phenomological model (Section 2.3.2.4).

2.2.4.3 Fouling amelioration

Fouling can suppressed in three ways: (a) pretreatment or in-treatment (i.e. membrane cleaning) to remove foulants; (b) promotion of turbulence to limit the thickness of the hydrodynamic boundary layer; and (c) reduction of the flux. Since all of these options add to the overall cost, either ostensibly operational (b) or capital (a, c), it is essential to optimise the system so as to suppress fouling, or ameliorate problems introduced by it, as much as possible without

adding excessively to the cost. In MBRs it is not feasible to remove the foulants by pretreatment, since it is these constituents which form a large part of the organic load which the MBR is intended to treat. Of the two remaining options, turbulance promotion is achieved by operating relatively wide bore of channel membrane elements, placed external to the bioreactor, at high crossflow. Flux reduction is employed for MBR systems in which the membrane is submerged in the bioreactor itself, thereby limiting the degree of turbulence promotion possible. It is in this regard that the so-called 'limiting' or 'critical flux' concept is considered.

2.2.4.4 The critical flux concept

Fouling can be considered to be either reversible or irreversible. A long term diminution in flux which is not recovered by simple cleaning techniques is indicative of irreversible fouling, and this is often attributable to colloidal deposition onto the membrane. Since mass transport of colloidal material to the membrane surface is directly related to the flux for any one membrane separation system, the flux at which colloidal deposition takes place is referred to the critical flux (Howell, 1995). Below this critical value the flux is directly proportional to transmembrane pressure and steady state operation is maintained. The critical flux is a function of the hydraulic conditions, tending to increase with increasing crossflow, and the nature of both the membrane and the polluting species.

Corroboratory evidence for the existence of a critical flux in aerobic MBR operation has been provided by Defrance and Jaffron (1999) using a TiO_2/ZrO_2 plate-and-frame ceramic membrane-based side-stream MBR. These authors demonstrated that beyond a sharply-defined value of the fixed operating flux of around 90 l m^{-2} h^{-1} and a crossflow velocity of 3 m s^{-1} the TMP increased dramatically with time, whereas below this value the TMP was constant. The critical flux was a linear function of crossflow velocity. During sub-critical operation of the MBR process non-fouling, low energy operation can be sustained resulting in greatly reduced cleaning requirements and savings in operational costs.

2.3 THE THEORY

There are essentially two approaches that can be employed to describe mass transport in membrane processes. One is to simply add the hydraulic resistance offered by the membrane to that offered by the cake or fouling layer in order to determine the flux through both matrices under a given pressure, or vice versa. This relies on a knowledge of the resistance of both membrane and fouling/cake

layer. However, provided the latter can be measured empirically and simple physical laws governing flow through porous media can be applied, this simple resistance theory approach can be usefully applied without recourse to further theoretical development. It is, indeed, common practice to refer to the membrane and cake resistance when defining dead-end filtration operation.

To develop predictive models for membrane processes from first principles, however, relies on mathematical description of the system hydrodynamics. A number of models have been presented to define the operational determinants of various membrane processes, and many of these are reviewed elsewhere (Fane, 1986; Davis, 1992; Lojkine *et al.*, 1992; Belfort *et al.*, 1994). To describe each of these in detail would be beyond the scope of this review since the outcomes, i.e. the ultimate analytical expressions generated, are specific to the process under consideration and the assumptions made. However, most of the mathematical developments referring to crossflow operation have their basis in film theory (also referred to as the concentration polarisation model). Classical film theory assumes diffusive transport in the interfacial region to be determined by the degree of concentration polarisation, which can then be calculated through a consideration of the system hydrodynamics.

2.3.1 Membrane mass transfer control

Under the simplest operational conditions, the resistance to flow is offered entirely by the membrane. For porous membrane systems, the flux can be expressed as:

$$J = \frac{\Delta p}{\mu R_m} \tag{4.5}$$

where J is the flux in m s^{-1}, Δp is the trans-membrane pressure, μ the fluid viscosity and R_m is the resistance of the membrane in units of m^{-1}. For microporous membranes, specifically those used for microfiltration, the Hagen-Poiseuille equation may be considered applicable for permeate undergoing laminar flow through cylindrical pores. The resistance R_m then equates to:

$$R_m = \frac{K(1 - \varepsilon_m)^2 S_m^2 l_m}{\varepsilon_m^3} \tag{4.6}$$

where ε is the porosity (or voidage), S_m the pore surface area to volume ratio and l_m the membrane thickness. K is a constant equal to 2 for perfectly cylindrical pores but changes for other geometries.

2.3.2 Cake layer mass transfer control

2.3.2.1 Resistance model

The simplest way of accounting for the additional resistance offered by the material accumulating in the interfacial region is to simply add the resistance R_c, of the cake layer to that of the membrane. Equation 4.5 then becomes:

$$J = \frac{\Delta p}{\mu(R_m + R_c)} \tag{4.7}$$

Under dead end operating conditions where (a) all suspended solids contributing to R_c are rejected by the membrane and (b) the bulk hydraulic resistance of the cake does not change with time, R_c is linearly related to the filtrate volume passed. Under such conditions, R_c follows the same form as Equation 4.6 and is represented by the Kozeny-Carman equation:

$$R_c = \frac{K'(1 - \varepsilon_c)^2 S_c^2 l_c}{\varepsilon_c^3} \tag{4.8}$$

where the symbols refer to the same parameters as before with reference to the filter cake. In this case, however, K' takes a value of 5 for spherical (or neo-spherical) geometry.

For crossflow operation, on the other hand, the resistance may be expected to attain a constant steady-state value once the adhesive forces retaining cake or fouling layer at the membrane surface are balanced by the shear forces acting at or near the hydrodynamic boundary layer. Thus, if there is sufficient information to calculate the hydraulic resistance of the cake and/or fouling layer, either from empirical measurement or from Equation 4.8, the steady-state flux can be calculated from Equation 4.7. However, in practice the flux in crossflow filtration systems invariably decays with time due to changes both in the bulk properties of the cake and in the membrane itself.

Notwithstanding its limitations, simple resistance theory forms the basis of classical cake filtration theory for dead-end operation, and many investigators choose to report results from practical work in terms of the hydraulic resistance values. For crossflow operation, however, a first-principles definition of hydraulic resistance can, in theory, be derived through a consideration of the balance of forces at the membrane:solution interface. This leads to a definition of classical, Brownian diffusion-driven concentration polarisation.

2.3.2.2 Classical concentration polarisation model

Concentration polarisation describes the tendency of the solute to build up in the membrane solution interfacial region, and the extent to which this occurs depends on:

- the propensity of the rejected (solute[1]) species to diffuse (i.e. their diffusivity)
- the notional thickness of the stagnant region
- the rate at which solute species are added to the stagnant region

The first of these is a property of the solute itself, and is related principally to its size. The latter two factors, on the other hand are determined mainly by the operating conditions themselves. The thickness of the stagnant region, normally denoted δ, can be determined from the rheological properties of the liquid, the prevailing hydrodynamic conditions and the dimensions of the flow channel. The rate at which ions arrive in the stagnant film is simple determined by the flux and the rejection. Therefore, provided the system is well defined the degree of concentration polarisation can be calculated and its effects on the operation of the membrane process assessed.

The mathematical description proceeds by conducting a material balance at the membrane, where the build up of solute at the interface is countered by the diffusive flux of solute away from membrane. Assuming a one-dimensional system (i.e. no longitudinal mass transfer), near 100% rejection and a constant value δ for the boundary layer thickness (Figure 2.4), the concentration polarisation under steady state conditions based on film theory can be defined as:

$$J = \frac{D_B}{\delta} \ln\left(\frac{C^*}{C}\right) \tag{4.9}$$

where D_B is the Brownian diffusion coefficient in $m^2\ s^{-1}$ and C^* and C are the respective concentrations at the membrane surface and in the bulk solution: the ratio D/δ represents the mass transfer coefficient k in units of $m\ s^{-1}$. Note that Equation 4.9 includes no pressure term, though the trans-membrane pressure is inferred by the flux value.

Determination of the flux from Equation 4.9 relies on knowledge of the solute diffusivity, the boundary layer thickness and the solute concentration at the membrane surface. If the solute comprises dissolved ions or small

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[1] the term 'solute', for the purposes of this discussion, includes suspended and dissolved rejected material

molecules, as would be the case for pressure-driven dense membrane processes, then D is simply given by the Stokes Einstein equation:

$$D_B = \frac{2\kappa T}{3\pi\mu r_p}$$ (4.10)

where κ is the Boltzmann constant, T is absolute temperature and r_p is the solute radius. δ is dependent only on the system hydrodynamics which, provided the fluid displays Newtonian behaviour, is simply a function of:

- physical properties of the liquid, which for most water treatment processes change only marginally with chemical water quality and can normally be accurately expressed as a function of temperature;
- the shape and size of the flow channels within the module; and
- the mean velocity and, in particular, the shear rate of liquid flowing through the channels.

This leads to the so-called Lévêque solution for laminar flow and Brownian diffusive transport (Lévêque, 1928; Porter, 1972), whereby the length-averaged flux is given by:

$$\langle J\rangle = 0.0807\left(\frac{D_B^2\gamma_0}{L}\right)^{1/3}\ln\left(\frac{C^*}{C}\right)$$ (4.11)

where γ_0 is the maximum shear rate and L the hydraulic dimension. The shear rate is given by the ratio of the crossflow velocity U to the characteristic dimension and is geometry dependent, thus for parallel flow channels of height h:

$$\gamma_0 = \frac{6U}{h}$$ (4.12)

and for tubes of diameter d:

$$\gamma_0 = \frac{8U}{d}$$ (4.13)

2.3.2.3 Modified concentration polarisation models

The Lévêque solution is based on channel flow, with the boundaries being completely impermeable. As such it is strictly only applicable to membrane permeation systems if the permeate flux is much smaller than the crossflow

velocity. Complications arise in the concentration polarisation model when it is applied specifically to systems in which colloidal and/or suspended material is present and accumulate in the hydrodynamic boundary layer. In such cases Newtonian behaviour cannot be assumed within or at the outer boundary of the stagnant film, and the deviation from classic film theory increases with increasing solute concentration in the boundary layer. This implies that a correction for non-Newtonian behaviour is needed to account for the local solute concentration-dependent changes in: viscosity of the fluid,, diffusivity of the solute, and permeability of the cake.

The various analytical expressions developed to describe non-classical behaviour are summarised in Table 2.7. The exact solution for the equilibrium flux varies according to the approach taken and the assumptions made. However, the general trend in the modified expressions is for transport of solute away from the membrane to be much higher than that predicted by Brownian diffusion. This then means that the nature of the dependency of flux on both crossflow velocity (which is directly proportional to shear rate) and particle size changes significantly from the Brownian diffusion Lévêque model if either shear-induced diffusion or inertial lift are accounted for. For example, dependency on crossflow changes from $U^{0.33}$ proportionality to direct proportionality for shear-induced diffusion (Zydney and Colton, 1986) or U^2 for inertial lift (Drew *et al.*, 1991). Kim and Park (1999) have based their prediction of critical flux conditions on shear-induced diffusion.

Perhaps the most comprehensive, and commensurately complex, solution for equilibrium flux in CFMF is that offered by Romero and Davis (1988). This model accounts the effects of shear-induced diffusion on a non-uniform filter cake whose thickness increases with axial membrane (channel) distance. This is considerably more complex than the model for thin cake deposits (Zydney and Colton, 1986), where a uniform cake deposit over the whole membrane area is envisaged, and requires that the solute concentration dependency of viscosity and diffusivity be predetermined. More recently, the Romero and Davis model has been slightly simplified by basing the cake layer resistance on the Kozeney Carman equation (Ould-Dris *et al.*, 2000).

Table 2.7:   Governing equations: steady-state expressions for the length averaged permeate flux

Model	Equation	Reference
Lévêque solution: laminar flow, Brownian diffusive transport, J<<U	$\langle J \rangle = 0.807\left(\dfrac{D_B^2 \gamma_o}{L}\right)^{1/3} \ln\left(\dfrac{C^*}{C}\right)$	Porter (1972), after Lévêque (1928)
Similarity solution for laminar flow, Brownian diffusive transport, J→U	$\langle J \rangle = 1.31\left(\dfrac{D_B^2 \gamma_o C^*}{CL}\right)^{1/3}$	Davis and Sherwood (1990) Romero and Davis (1988)
Fully-developed turbulent flow	$\langle J \rangle = 0.023\left(\dfrac{U^{0.8} D_B^{0.67}}{d^{0.2} v^{0.47}}\right)^{1/3} \ln\left(\dfrac{C^*}{C}\right)$	Porter (1972)
Lévêque solution for shear-induced diffusion (based on $D_s = 0.03 r^2 \gamma_o$)	$\langle J \rangle = 0.078\left(\dfrac{r^4}{L}\right)^{1/2} \gamma_o \ln\left(\dfrac{C^*}{C}\right)$	Zydney and Colton (1986), after Ecstein et al (1977)
Similarity solution for shear-induced diffusion (based on C*~0.6 by volume and C<0.1 by volume)	$\langle J \rangle = 0.072 \gamma_o\left(\dfrac{r^4 C^*}{CL}\right)^{1/3}$	Davis and Sherwood (1990)
Integral model for shear-induced diffusion from thick layers (based on $D_s(C)$)	$\langle J \rangle = 1.31 \gamma_o \underline{\eta(C)}\left(\dfrac{r^4 \underline{Q_{er}(C)}}{LC}\right)^{1/3}$	Romero and Davis (1988)
Inertial lift velocity (based on thin layers, such that J=v_L	$J = \dfrac{0.036 r^3 \gamma^2}{16 v_o}$	Drew et al (1991)
Surface transport	$\langle J \rangle = 2.4 r \gamma_o \left(r^2 R_c'\right)^{2/5} \cot \theta$	Sherwood (1988)

Symbols

D_B, D_S	Diffusion coefficient for Brownian and shear-induced diffusion respectively
γ_o	Maximum shear rate (Equations 12 and 13)
C^*,C	Concentration at the membrane:solution interface and the bulk retentate solution respectively
$\underline{C}$	Dimensionless concentration
L	Membrane elemental length
U	Crossflow velocity
υ	Kinematic viscosity (η/ρ)
r	Particle radius
$\underline{\eta}(C)$	Concentration-dependent dimensionless viscosity
$Q_{er}(C)$	Concentration-dependent *excess particle flux*
R_c'	Specific cake resistance (R_c/cake thickness)
v_L	Inertial lift velocity
cot θ	Surface morphology parameter

Table 2.8: Governing equations: expressions for dynamic behaviour

Model	Equation	Reference
Pore blocking model	$J = J_o \exp\left(\dfrac{-a_{bl} J_o A}{n_p}\right) t$	Hermia (1982)
Pore constriction model	$J = \dfrac{J_o}{\left(1 + \dfrac{a_{pore} J_o}{n_p \pi \delta_m r_p^2} t\right)^2}$	Hermia (1982)
Cake formation model	$J = \left(1 + \dfrac{2 a_{dep} R_c' C_b A \Delta P}{\eta_o R_m^2} t\right)^{-1/2}$	Hermia (1982)
Phenomenological (protein)	$J = \dfrac{\Delta P}{\eta_o (R_m + R_c' M_c^* (1 - e^{-k_1 t}) + R_{bl}' M_{bl})}$	Fane (1986)
Phenomenological (activated sludge)	$J = \dfrac{\Delta P}{\eta_o (k_2 U^{-2.5} + k_3) R' U}$	Fane (1986)
Shear-induced diffusion from thick layers	$J = J_o \left(1 + \dfrac{2 J_o R_c' C}{R_m (C_c - C)} t\right)^{-1/2}$	Romero and Davis (1988)
Brownian diffusion from thick layers	$\langle J \rangle = 1.31 \left(\dfrac{D^2 \gamma_o}{L}\right)^{1/3} \left(\dfrac{C_c}{C} - 1\right)^{1/3}$ for $t \geq t_{ss}$ $\langle J \rangle = \dfrac{1}{L}\left[\int_0^{X(t)} J_{ss}(x)dx + [L - X(t)]J(t)\right]$ for $t < t_{ss}$ $X(t) = 4.81(D^2 \gamma_o)\left(\dfrac{C_c}{C} - 1\right)\left(\dfrac{C}{C_c}\right)^{3/2}\left(\dfrac{R_c' t}{\Delta P - \Delta P_c}\right)$ $\Delta P_c = \dfrac{3kT}{4\pi r^3} N_F$ $J(t) = \left(\dfrac{\Delta P - \Delta P_c}{R_m}\right)\left(1 + \dfrac{2 R_c'(\Delta P - \Delta P_c)}{R_m^2} \dfrac{C}{C^*} t\right)^{-1/2}$	Song (1998), after Song and Elimelech (1995)

a_{block}	No. pores blocked per filtrate volume passed
a_{pore}	Foulant volume deposited within pores per filtrate volume passed
a_{dep}	Fraction of foulant depositing on membrane
A	Filter area
J_o, J, J_{ss}	Flux initially, at time t, at equilibrium
$k_{1,2,3}$	Empirical first-order rate constants
M_{bl}	Boundary layer deposit
M_c^*	Maximum cake deposit (mass per unit area)
n_p	No. pores
r_p	Pore radius
R_m	Membrane resistance
R_{bl}'	Specific boundary layer resistance
ΔP	Operating pressure difference
ΔP_c	Critical pressure difference: pressure required for cake to form
t	Filtration time
t_{ss}	Steady-state filtration time: time after which flux is constant
X	Distance of the front of the dynamic layer from the entrance
η_o	Bulk suspension viscosity

2.3.2.4 Dynamic modelling

Solutions for models describing flux decline in dead-end filtration via the three main mechanisms for fouling, these being pore blocking, pore restriction and cake formation (Section 2.2.4.2) all demand experimental determination of the key parameter denoted 'a' in the equations listed in Table 2.7. These three models are strictly speaking only applicable to dead-end operation, in which it can be assumed that all the solute in the feed ends up on the membrane, but the models can all be adapted for crossflow operation if the proportion of undeposited solute material can be calculated.

Several phenomenological expressions have been presented for different systems, two of which are presented in Table 2.7 and include activated sludge filtration. While these equations can provide a useful description of dynamic behaviour they are unlikely to be universally applicable for highly heterogeneous matrices such as activated sludge. Indeed, most recent publications of experimental studies of MBR processes make little or no reference to dynamic modelling, with data interpretation being substantially limited to reporting of hydraulic resistance values (Choo and Lee, 1996; Chang and Lee, 1998). The more global semi-empirical models that have been developed (Nagaoka *et al.*, 1998), like most other semi-empirical models in this area, rely on specific hydraulic resistance data which is likely to be specific to the system under investigation.

2.3.2.5 Practical verification of modified models

Experimental studies on model colloidal or particulate systems have shown close agreement between experimentally measured steady-state flux data and those predicted from shear-induced diffusive mass transport theory for ideal systems, such as latex, blood, bacterial and fractionated clay suspensions (Zydney and Colton, 1986). The model of Kin and Park (1999) for predicting critical flux, again based on shear-induced diffusion, appears to corroborate well with experimental data from calcium carbonate filtration, the critical flux increasing linearly with particle size. Inertial lift, on the other hand, would appear to be restricted in importance to high shear rates and/or large particles (Davis, 1992).

The shear-induced diffusion model for thick cake layers (Romero and Davis, 1988, 1990) was found to give a reasonable representation of both dynamic and steady-state behaviour for both rectangular channels and ceramic tubular membranes challenged with homodispersed spheres of 0.45–1.37μm (Romero and Davis, 1991). Ould-Dris *et al.* (2000), using a slightly simplified adaptation of the Romero and Davis model, also found reasonable agreement between theoretical and experimental steady-state flux for their trials on the less idealised

system of granular calcium carbonate. Agreement was only obtained, however, after allowance was made for differing mean particle size of the cake layer and the bulk suspension, which required experimental verification. A similar limitation appears to apply to the Song model (Song and Elimelech, 1995): an excellent fit with theoretically predicted flux was obtained for CFMF of $0.06\mu m$ homodispersed colloidal silica once the specific cake resistance R'_c and the cake concentration C_c had been surmised (Wang and Song, 1999).

It is generally the case that the modified concentration polarisation models developed to define dynamic and steady state behaviour during microfiltration are very sensitive to key parameters pertaining to particle transport and the hydraulic resistance of the cake. These parameters are either only calculable for highly idealised systems or else must be determined empirically. For a highly complex matrix such as municipal wastewater and/or a hydrodynamically complex system such as a submerged MBR, the useful employment these models for predictive purposes is probably restricted to defining trends. Moreover, there appears to be a paucity of mechanistic information on fouling and dynamic behaviour of submerged MBR systems that could be used to form the basis of a predictive model.

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3

Fundamentals of biological processes

3.1 INTRODUCTION

Biological processes are primarily designed for the removal of dissolved and suspended organic matter from wastewaters. The correct environmental conditions are provided to encourage the growth of micro-organisms which use the organic compounds, often measured as biochemical oxygen demand (BOD) or chemical oxygen demand (COD), as carbon substrate. The micro-organisms that grow on the substrate are subsequently separated from the water which has had the BOD removed, leaving a relatively clean effluent. They derive energy and cellular material from the oxidation of this organic matter and can be aerobic or anaerobic. Biological wastewater treatment is also capable of removing other wastewater components, including suspended solids, nitrogen, phosphorus, heavy metals and xenobiotics.

[41]

One advantage of using biological processes is that they are seen as 'natural': the reactor systems merely intensifying processes that might occur in the environment. Soluble and solid wastes are transformed, being converted to gases, either carbon dioxide if aerobic or carbon dioxide and methane if anaerobic, inert solids and water. The methane can be used as an energy source. The waste solids, termed sludge, are treatable prior to final disposal and odours are usually reduced. With most processes, high removal efficiencies are possible and variable loads can be tolerated. However, biological processes are susceptible to toxic chemicals and slow compared to chemical treatment. Soluble material generates solids that need to disposed of and can produce noxious compounds. Finally, many aerobic processes are net energy consumers, particularly aerobic suspended growth systems.

3.2 PROCESS TYPES

Biological processes can be classified by feeding regime, i.e. whether they are continuous or fed-batch; redox conditions; and whether fixed film or suspended growth.

As with most processes, if used for large volumes and continuous flows, biological treatment will be operated receiving a continuous flow. If smaller volumes are involved, such as for sludge treatment or industrial wastewater treatment, fed-batch processes can be used. Sequencing batch reactors (SBRs) are fed-batch activated sludge systems that use the same tankage for the bioreactor and sedimentation (Irvine and Ketchum, 1989). This means that the overall capital cost can be reduced and treatment of continuous flows will require at least two units operating in parallel. The sequence begins with the tank only partly filled and aeration off so that influent can be received. Aeration is continual for the 'react' phase, followed by quiescent settling. The supernatant is then withdrawn, being equivalent to final effluent. Other phases can be inserted, e.g. no aeration, merely mixing, as the tank is filled. Membrane bioreactors have usually been operated as continuous processes (Brindle and Stephenson, 1996) but there have been reports of fed-batch systems (Wilderer et al., 1985).

Redox conditions are usually categorised as aerobic, anoxic and anaerobic. Treatment systems designed for organic carbon removal are either aerobic or anaerobic; anoxic systems are usually designed primarily for nitrogen removal. In both aerobic and anoxic systems, oxygen is the final electron acceptor: this is provided by dissolved oxygen in the former and nitrate oxygen usually in the latter. Anaerobic processes occur in the complete absence of oxygen. In general, anaerobic catabolism produces less energy than aerobic catabolism, therefore

larger reactor volumes are required. Advantages and disadvantages of the two processes are listed below in Table 3.1. The main three types of MBR processes, biomass separation, extractive and bubble-less gas transfer, have been used in combination with aerobic biology (Brindle and Stephenson, 1996). Membrane bioreactors with anoxic stages have been used for nitrate removal (Suwa *et al.*, 1992). Anaerobic processes are suitable for high strength wastewaters and a system using membrane separation has been applied at full-scale treating maize processing wastewaters (Ross *et al.*, 1992).

Table 3.1: Anaerobic versus aerobic systems

	Anaerobic	Aerobic
Energy	low	high
% removal	60–90	95+
Sludge production	low	high
Stability	low–moderate	moderate–high
Start-up	2–4 months	2–4 weeks
Odour	potential	less
Alkalinity	high	Low
Biogas	yes	no
Nutrients	Low	can be high

Suspended growth processes rely upon the microbes (biomass) being in free suspension, as in the activated sludge process. Contact with the wastewater constituents will rely upon good mixing in such reactors. One advantage of suspended growth processes is the ability to more closely control the biomass retention time (see section 3.4 below). Fixed film reactors provide an inert support material on which the micro-organisms can grow. The microbial population is then in contact with the wastewater as it is passed over the support matrix. It is important to note that with both types of systems a separation stage is needed to remove the biomass from the clean effluent, usually sedimentation. Almost all bioreactor systems using membranes for biomass separation are suspended growth (Brindle and Stephenson, 1996), including the main aerobic systems applied at full-scale (Stephenson and Brindle, 1999). Fixed film systems form the basis of bubble-less gas transfer MBRs (Pankhania *et al.*, 1994) and also have a role in extractive MBR systems (Livingston, 1994). The different combinations of systems are summarised in Table 3.2 with examples of MBRs where they have been reported.

Table 3.2: Classification of biological wastewater treatment processes with examples of MBRs

Process	System	Reactor	Function	Configuration	Membrane Bioreactors
Aerobic	suspended-growth	continuous	BOD removal nitrification	activated sludge oxidation ditch contact stabilisation aerobic pond treatment	Chiemchaisri and Yamamoto 1993 Muller et al., 1995 Ciesk et al., 1999
Aerobic	suspended-growth	fed-batch	BOD removal nitrification	sequencing batch reactor	Wilderer et al., 1985
Aerobic	fixed film	continuous or fed-batch	BOD removal nitrification	trickling filtration high-rate filtration rotating biological contactor	Brindle and Stephenson 1996
Aerobic	suspended-growth or fixed film	continuous	BOD removal nitrification	biological aerated filtration trickling filter solids-contactor process	
Anoxic	suspended-growth or fixed film	continuous	nitrification denitrification	tertiary denitrification	Gunder and Krauth 1999
Anaerobic	suspended-growth	fed-batch	Solids reduction	sludge digestion	
Anaerobic	suspended-growth	continuous	BOD removal	contact process sludge blanket reactor anaerobic pond treatment	Ross et al., 1992
Aerobic/ Anoxic	suspended-growth or fixed film	continuous	BOD removal nitrification denitrification	multi-stage activated sludge filter	Buisson et al., 1998 Yoon et al., 1999
Aerobic/ Anoxic/ Anaerobic	suspended-growth	continuous	BOD removal nitrification denitrification phosphate removal	multi-stage activated sludge	

3.3 MICROBIOLOGICAL FUNDAMENTALS

Biological treatment systems rely upon many types of micro-organisms, being present in the same reactor. Bacteria have the key roles which includes conversion of soluble and particulate organic compounds into biomass and gaseous waste products, conversion of ammonia to nitrate (nitrification) and conversion of nitrate to nitrogen gas (denitrification). Higher forms of micro-organisms, e.g. protozoa and rotifers, play crucial roles in consuming particulate organics, including scavenging of bacteria. Other larger biological species such as nematode worms and insect larvae may contribute to the consumption of particulate organic matter, especially in trickling filter systems. Cicek *et al.* (1999) reported few filamentous organisms, nematodes and ciliates in MBRs operated at 30 d sludge age compared to activated sludge operated at 20 d sludge age. Variations in conditions such as shear stress, mass transfer, mixing and absence of a clarifier were cited as reasons. In contrast, Ghyoot and Verstraete (2000) noted higher concentrations of protozoa, particularly flagellates and free ciliates, in a submerged MBR when compared to activated sludge operating at the same sludge age. Some of the degradation in a biological process is undertaken by extracellular as well as intracellular enzymes. Enzymatic analysis of MBR and activated sludge biomass demonstrated that overall and soluble phase activity was higher in the MBR (Cicek *et al.*, 1999).

Micro-organisms can be classified according to the redox conditions in which they survive (thereby determining process design to encourage their growth—see 3.2 above) and their the carbon and energy requirements. Heterotrophs use organic carbon as an energy source and as the carbon source for synthesis of more cellular material (Table 3.3). Those bacteria in engineered treatment processes responsible for BOD removal and denitrification are heterotrophs. Autotrophs use inorganic reactions to derive energy, e.g. oxidation of iron (II) to iron (III), and obtain carbon from an inorganic source, e.g. carbon dioxide. Bacteria responsible for nitrification, sulphate reduction and anaerobic methane formation are autotrophs. As a general rule, autotrophs are less efficient at energy gathering than heterotrophs and therefore grow slower.

Table 3.3 Microbial metabolism types used in biological wastewater treatment processes

Component	Process	Electron Acceptor	Type
Organic-carbon	aerobic biodegradation	O_2	aerobic
Ammonia	nitrification	O_2	aerobic
Nitrate	denitrification	NO_3.	facultative
Sulphate	sulphate reduction	SO_4^{2-}	anaerobic
Organic-carbon	methanogensis	CO_2	anaerobic

Important environmental conditions for microbial growth that need consideration are total dissolved solids (TDS) concentration, pH and temperature. Most micro-organisms can only function in relatively dilute solutions, around neutral pH and at ambient temperatures. Some types are able to grow under extreme conditions e.g. *Thiobacillus* growth optimum is at pH 1.5 to 2.0, and can be used in specialised biological reactors. Membrane systems have been linked to bioreactors to enable the biodegradation of organic components in waste streams in which survival of micro-organisms is impossible. The extractive MBR allows for the removal of chlorinated organic compounds from waste streams of pH<1 and high TDS (Livingston, 1994). Different micro-organisms have different temperature profiles and can be classified as psychrophilic, mesophilic and thermophilic with optimum growth at temperatures of 15, 35 and 55 °C respectively. While most aerobic biological processes are operated at ambient temperatures, the micro-organisms usually have mesophilic temperature optima. Van Dijk and Roncken (1997) noted that in addition to the heat of biological reaction, the energy input of crossflow ultrafiltration could raise temperatures in MBRs to the optimum 35 °C.

3.4 KINETICS AND PROCESS OPERATION

The design and behaviour of biological processes can be characterised by considering the kinetics of microbial growth. This is described by using Monod kinetics, i.e. the rate of reaction is first order with respect to a limiting substrate up to a maximum specific growth rate, after which point the growth rate is unaffected by any increase in substrate concentration:

$$\mu = \frac{\mu_m S}{K_s + S} \qquad (3.1)$$

where μ is the growth rate (h^{-1}) μ_m is maximum specific growth rate (h^{-1}), K_s is the saturation coefficient and S is substrate concentration (kg m^{-3}). The limiting

substrate in most wastewater treatment systems will be organic carbon, i.e. BOD/COD. Monod kinetics have been the underlying basis for describing microbial growth in MBRs (e.g. Chaize and Huyard, 1991; Wen *et al.*, 1999).

To describe kinetics that would be useful for understanding and designing biological wastewater treatment processes, a suspended growth culture in a stirred tank reactor which is continuously fed with substrate needs to be considered. Detailed considerations of kinetics relevant to biological wastewater treatment can be found in texts such as Metcalf and Eddy (1991) and Grady *et al.*, (1999). Under steady-state conditions a mass balance combined with Monod kinetics gives:

$$S = \frac{K_s D}{\mu_m - D} \qquad (3.2)$$

where D is dilution rate (h^{-1}), i.e. the inverse of hydraulic retention time (HRT). Thus it can be seen that up to the maximum specific growth rate for a given influent concentration, a change in HRT will alter the exit substrate concentration. However, before exit substrate can be predicted in real systems, endogenous metabolism, i.e. cells utilizing stored materials and extracellular polymers associated with the biomass, also has to be taken into account. This endogenous metabolism is accounted for in the mass balance for the reactor by the 'death' rate constant (k_e). Therefore the exit substrate concentration (S_e) is described by:

$$S_e = \frac{K_s (D + k_e)}{[\mu_m - (D + k_e)]} \qquad (3.3)$$

Death rate constants for conventional activated sludge processes and anaerobic processes might typically be 0.05–0.06 d^{-1} at 20 °C (Metcalf and Eddy, 1991). Wen *et al.*, (1999) found k_e to be 0.08 d^{-1} at 30 °C in a ceramic membrane sidestream aerobic bioreactor treating raw wastewater. Fan *et al.*, (1996) quoted 0.05 d^{-1} for an MBR at the same temperature. If it is known how much biomass is produced for the substrate removed, this can be related to equation (3.3). The amount of biomass produced is described by the yield coefficient (Y). This is the ratio of biomass produced per unit mass of carbon substrate used per unit time. The sludge yield is an important parameter to determine the total amount of waste biomass produced in a biological process. Observed yields (Y_{obs}) are approximately 0.6 d^{-1} for conventional aerobic

processes and an order of magnitude lower for anaerobic ones. In the stirred tank reactor, biomass production can therefore be described by:

$$X = YD\frac{(S-S_e)}{(D+k_e)} \tag{3.4}$$

where X is biomass concentration (kg m^{-3})

These equations demonstrate that by controlling the HRT in a stirred tank reactor, the growth rate of the micro-organisms is controlled. By controlling growth rate, the treatment performance of a biological process is determined and the production of waste biomass can be predicted. Unfortunately, micro-organisms grow slowly relative to the rate of chemical reactions in dilute systems (which most wastewaters are); therefore in order to design many biological processes that are economic, long biomass retention times are needed. This can simply be achieved by using a very large reactor volume, which is applied in anaerobic sludge digestion. Otherwise, uncoupling the solids retention time from the HRT by using fixed film systems, i.e. the microbes are attached to an inert media, can reduce reactor volume. Alternatively, suspended growth systems coupled to a sedimentation process with biomass recycle to the reactor can be used, i.e. activated sludge-type processes. In this type of system, the growth rate of the micro-organisms can be controlled independently of the HRT through wastage of the sludge (Fig. 3.1).
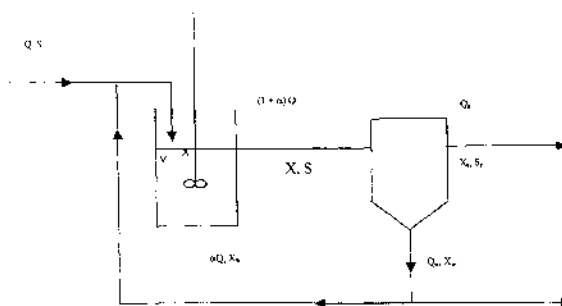
Figure 3.1. Schematic diagram of cell culture with sedimentation and recycle - the activated sludge process.

Therefore the removal of biomass, i.e. control of the average specific growth rate of the population of micro-organisms, is undertaken deliberately in

engineered systems and contributes to control of overall performance. This is termed the sludge age (θ_x) and can be calculated as follows:

$$\theta_x = \frac{VX}{Q_w\,X_w + Q_c\,X_c} \qquad (3.5)$$

Where V is the aeration tank volume (m^3, Q_w is the sludge wastage rate ($m^3\,d^{-1}$), X_w is the suspended solids concentration of the waste sludge ($kg\,m^{-3}$), Q_c is the effluent flow rate ($m^3\,d^{-1}$) and X_e is the effluent suspended solids concentration ($kg\,m^{-3}$). Therefore controlling the sludge age should control the final effluent quality of the activated sludge process. Unfortunately, this is more dependent on a factor that is not predictable: the settleability of the sludge.

An important empirical design parameter used for the design and control of activated sludge is the food-to-microorganism ratio (F:M):

$$F:M = \frac{S\,Q}{VX} \qquad (3.6)$$

where Q is the influent flow rate ($m^3\,d^{-1}$).

The term ratio is a misnomer; as the units are d^{-1}, F:M is more analogous to a rate constant. The F:M is ultimately controlled by the sludge age and can be related to it by:

$$\frac{1}{\theta_x} = Y(F:M)\frac{E}{100} - k_e \qquad (3.7)$$

where E is process efficiency (%).

Sludge ages for activated sludge plants treating municipal wastewaters are typically in the range of 5 to 15 d, with F:M of 0.2 to 0.4 d^{-1}. At long sludge ages, the biomass concentration maintained in the reactor, termed mixed liquor suspended solids (MLSS), is higher. Conventional systems, operating at sludge ages of ca. 8 d treating municipal wastewater, will have an MLSS of ca. 2,500 mg^{-1}, whereas an activated sludge plant with a sludge age of ca. 35 d might have a MLSS of 8,000 mg l^{-1}. One potential advantage of biomass separation MBRs is the ability to operate at high sludge ages, and therefore high MLSS and a low F:M, thereby reducing sludge yield.

Of the many ways of reducing sludge yield in aerobic systems (Mayhew and Stephenson, 1997), the most simple in suspended growth systems is to increase

the sludge age, i.e. reduce the wastage rate (Q_w in equation 3.5). The organic substrate is used by micro-organisms for two things; synthesis of more biomass; and for cell maintenance. The latter results in production of waste gases – methane and carbon dioxide if anaerobic, nitrogen and carbon dioxide if anoxic, and carbon dioxide if aerobic. Therefore the higher the yield coefficient, the more biomass and less carbon dioxide is produced from degradation of the substrate. The observed yield (Y_{obs}) is lower than the actual yield (Y) as a result of endogeneous respiration (k_e). This mode of operation is explained in terms of the maintenance concept as described by Pirt (1975). The maintenance concept describes operation where all incoming substrate is used for cell maintenance rather than growth, such that no excess sludge is produced. This requires a condition of substrate limitation in the reactor brought about through a long sludge age resulting in a low F:M ratio. Membrane bioreactors have been used to study the maintenance concept using single cultures of *Pseudomonas fluorescens* (Bouillot *et al.*, 1990). It was observed that the maintenance coefficient was similar for both growing (0.035) and zero growth (0.042) systems. Canales *et al.*, (1994) also undertook a detailed study of the maintenance phenomena using cultures of *Pseudomonas fluorescens* in an MBR. It was observed that under normal operating conditions, sludge yields were 0.56 kg kg^{-1} COD. When raw cell lysis products and the soluble fraction of the cell lysis products were added to mimic the situation with significant endogenous matabolism, the yield was 0.36 and 0.22 kg kg^{-1} COD respectively. This demonstrated that at high sludge ages, endogenous metabolism resulted in a decreased yield.

Muller *et al.*, (1995) operated a membrane bioreactor for 300 d without wasting any sludge, i.e Y_{obs} was close to zero. The MLSS reached almost 50,000 mg l^{-1} towards the end, when the oxygen transfer rate became poor. Benitez *et al.* (1995) treated a pharmaceutical wastewater in an MBR and wasted no sludge in a 62 d experiment. The MLSS had stabilised at 25,000 mg l^{-1} at the end of the study. A pilot-scale submerged aerated MBR operated at a sludge age of 50 d and treating municipal wastewater had a sludge production rate of approximately 0.25 kgSS kg^{-1} COD removed (Cote *et al.*, 1997). The authors stated that this was 'about 50% less than that of an extended aeration activated sludge process'. Both Lubbecke *et al.*, (1995) and Davies *et al.*, (1998) observed that sludge production in MBRs was 70% less than for conventional activated sludge at the same space loading.

Membrane bioreactors can be operated in the same way as conventional activated sludge plants with a defined sludge age. Ghyoot and Verstraete (2000) operated a pair of two stage bioreactors treating a synthetic wastewater based on skimmed milk. The first stage in each case was an aerated, completely mixed reactor; the second stage for one system was a conventional activated sludge

reactor and for the other system the second stage was a submerged MBR. Both systems were operated at 3 sludge ages; at 102 d the maintenance effect was apparent with very low observed yields (Table 3.4)

Table 3.4. Sludge yield at different sludge ages with two-stage conventional activated sludge (AS) and MBR configurations (Ghyoot and Verstraete, 2000).

Sludge age (d)	12	24	102
AS yield (kgSS kg^{-1} COD removed)	0.28	0.26	0.07
MBR yield (kgSS kg^{-1} COD removed)	0.22	0.18	0.02

It has been hypothesized that under zero sludge wastage a gradual build up of non-volatile solids would occur and hence activity. Muller *et al.*, (1995) found that the non-volatile fraction remained relatively stable over 300 d at 20.5 to 23.6% of the sludge. Rosenburger *et al.*, (1999) demonstrated that over a continuous 3 year operation no decrease in the volatile fraction was observed with the MLVSS/MLSS ratio remaining constant at approximately 75% throughout.

3.5. AERATION

In conventional aerobic biological wastewater treatment processes oxygen is usually supplied as atmospheric air, either via submerged air-bubble diffusers or surface aeration. Diffused air bubbles are added to the bulk liquid (activated sludge, biological aerated filters, fluidised bioreactors, etc. or oxygen transfer occurs from the surrounding air to the bulk liquid via the liquid/air interface (as for biofilm processes such as trickling filters or rotating biological contactors). The oxygen required by a biological process to degrade a known amount of organic matter can be calculated from:

$$Q_o = OUE \cdot Q \ (S - S_e) + b \, VX \qquad (3.8)$$

where Q_o is oxygen requirement (kg d^{-1}), OUE is oxygen utilisation efficiency (dimensionless) and b is the endogenous oxygen demand coefficient (d^{-1}). It can be seen from this equation that in addition to the substrate exerting an oxygen demand, a higher MLSS will increase demand. High MLSS can be maintained in MBRs; indeed, Muller *et al.*, (1995) noted that at 50,000 mg l^{-1} MLSS, an MBR became oxygen transfer limited.

In order to satisfy the oxygen requirement, the gas has to be transferred into the liquid at a fast enough rate; this can be calculated from:

$$OTR = K_La(C^*-C) \qquad (3.9)$$

where OTR is oxygen transfer rate (kg m^{-3} d^{-1}), K$_L$a is the overall mass transfer coefficient (d^{-1}), a is the gas-liquid interface surface area (m^2), C* is the oxygen saturation concentration (kg m^{-3}) and C is the dissolved oxygen concentration (kg m^{-3}). The dissolved oxygen concentration in equilibrium with the oxygen partial pressure can be calculated from Henry's Law;

$$C^* = \frac{P_T \cdot \text{mole fraction of } O_2 \text{ in gas}}{H_C} \qquad (3.10)$$

where H$_C$ is Henry's constant (atm m^3 kg^{-1}) and P$_T$ is total gas pressure (atm).

As well as providing the oxygen required for microbial activity, aeration is also used for hydrodynamic mixing to ensure high mass transfer rates. A compromise must be made regarding the amount of aeration and the ideal bubble size required to satisfy this dual role. Consequently oxygen utilisation can be surprisingly low; typically 80 to 90% of oxygen diffused as air in the activated sludge process is lost to atmosphere. One measure of efficiency is the standard aeration efficiency (SAE) which can be calculated from:

$$SAE = \frac{OTR \cdot a}{\text{power requirement}} \qquad (3.10)$$

The SAE is a measure of the amount of oxygen transferred to the bulk liquid per unit of energy (kgO$_2$ kWh^{-1}). Air aeration limitations can be overcome by using oxygen-enriched air or high purity oxygen, the latter treatment increases the saturation concentration (C*) by approximately 4.7 times. Such processes have a greater volumetric degradation capacity compared to conventional air aeration processes. As a result of the high cost of oxygen, processes need to achieve high OUEs and SAEs. One way of increasing the efficiency is to transfer oxygen directly to biomass; this is the basis of the membrane aeration bioreactor (Brindle *et al.*, 1998) described fully in Chapter 5.

3.6   NUTRIENT REMOVAL

Under the right conditions the processes of nitrification and denitrification can be encouraged, i.e. the conversion of ammonia to nitrogen gas. Nitrification is the conversion of ammonia to nitrate by micro-organisms (Barnes and Bliss, 1983). This is achieved in two stages with the conversion of ammonia to nitrite by *Nitrosomonas* followed by conversion of nitrite to nitrate by *Nitrobacter*. The first step is rate limiting and therefore nitrite concentrations do not build up in most biological treatment systems as the *Nitrobacter* will immediately convert this compound to the nitrate. Both nitrifying organisms, being autotrophs, grow slower than most of the heterotrophic micro-organisms utilised during biological wastewater treatment. Fan *et al.*, (1996) found nitrifier growth rates of 0.1 to 0.2 d^{-1} in a ceramic MBR treating municipal wastewater; lower than the range of 0.28–2.2 d^{-1} quoted by Sharma and Ahlert (1977). In particular, as nitrifiers are slower growing than heterotrophs, longer sludge ages are required in order to achieve full (>90%) nitrification compared with BOD removal.

For nitrification to be successful, there needs to be sufficient concentrations of the three substrates: carbon dioxide, ammonia and oxygen. As autotrophs, carbon dioxide provides carbon for cell growth. Nitrifiers are obligate aerobes and dissolved oxygen (DO) concentrations need to be 1.0 to 1.5 mg l^{-1} in suspended growth systems for the survival of the bacteria. In biomass separation MBRs complete nitrification has been reported in sidestream systems with 3 mg l^{-1} DO (Trouve *et al.*, 1994). Chiemchaisri *et al.* (1992) noted inhibition of ammonia removal at 0.5 mg l^{-1} DO with recovery of full nitrification at 1.0 mg l^{-1}. Nitrification is also temperature sensitive, with decreased removal below 10 °C, which has also been observed in MBRs (Chiemchaisri and Yamamoto, 1993). Biochemical analysis confirmed a decrease in Q-8 ubiquinone found in strictly aerobic bacteria, indicative of a decrease in nitrifiers.

Nitrates can be removed from wastewaters through denitrification by bacteria that normally remove BOD under aerobic conditions but under anoxic conditions are able to convert nitrates to nitrogen gas, i.e. facultative micro-organisms. Wisniewski *et al.*, (1999) used an anoxic MBR to denitrify potassium nitrate for ethanol removal so as to determine the kinetics of organic carbon removal. Membrane bioreactors have been operated with an anoxic zone and achieved total nitrogen removal equal to that in a parallel activated sludge system (Yoon *et al.*, 1998).

As most wastewaters treated by biological processes are carbon limited, phosphorus is not removed to any great extent. Membrane bioreactors are no different: Yoon et al., (1999) noted that phosphorus removal was not improved in a MBR compared to a conventional system. Enhanced biological phosphate removal can be achieved by the addition of an anaerobic zone at the front of an activated sludge plant and returning nitrate-free sludge from the aerobic zone (Yeoman et al., 1986). There is no reason this could not be applied to an MBR system.

3.7 BIOMASS SEPARATION

Gravity settlement is usually used for separation of biomass from the final effluent—it is often the Achilles' Heel of biological processes. Biological aerated filters (BAFs) use depth filtration for separation of biomass from the final effluent (Mendoza-Espinosa and Stephenson, 1999). While the kinetic description presented in section 3.4 above allows for prediction of the concentration of substrate in the final effluent, it does not allow for accurate determination of final effluent BOD or suspended solids after sedimentation. The physical properties of the sludge and the configuration and operation of the sedimentation process will have a major impact. Sedimentation does not work well for anaerobic systems due to gas bubbles floating sludge to the surface. In aerobic systems, denitrification can occur resulting in nitrogen gas raising sludge to the surface. In bulking, filamentous micro-organism outgrow other bacterial species and produce less dense, bulky flocs that do not settle as. Membranes for biomass separation would avoid such problems.

Use of sedimentation for biomass separation will select for larger floc sizes compared with biomass separation MBRs. Zhang et al., (1997) compared four MBRs with four conventional activated sludge processes. The size distribution of flocs were smaller in the MBRs at 7–40 µm compared with 70–300 µm in activated sludge. Wisniewski and Grasmick (1998) demonstrated that the particle size characteristics changed with different rates of recirculation through a sidestream microfiltration module. From an initial mixed liquor seed from a conventional activated sludge plant the non-settleable fraction increased from only 1.5–3% of total COD to almost 100% after 150 h of operation. In a direct comparison of activated sludge and membrane bioreactors, Cicek et al., (1999) found that the former had a sludge volume index (SVI) that averaged 80 ml g^{-1} whereas the MBR sludge did not settle at all. In the MBRs, 97% of particles were <10 µm, with most of the surface area made up of particles in the range of 3–5 µm. In the activated sludge process, only 88% of particles were <10 µm with the main contribution to the surface area from particles in the size range 80–120 µm.

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4

Biomass separation membrane bioreactors

4.1 INTRODUCTION

Biomass separation membrane bioreactors, the most common type of MBR, are the amalgamation of a suspended growth reactor and membrane filtration device into a single unit process. The membrane unit can be configured external to, as in sidestream operation, or immersed in the bioreactor (Figure 4.1). Biomass separation MBRs should not be confused with the use of membranes for tertiary treatment after a biological process and sedimentation. In the case of an external system the membrane is independent of the bioreactor. Feed enters the bioreactor where it contacts biomass. This mixture is then pumped around a recirculation loop containing a membrane unit where the permeate is discharged and the retentate returned to the tank. The transmembrane pressure (TMP) and the crossflow velocity, which define the operation of the membrane, are both generated from a pump. Immersed systems differ in that there is no recirculation loop as the separation occurs within the bioreactor itself. Under these

circumstances the TMP is derived from the hydraulic head of the water above the membrane. In some systems this is supplemented by a suction pump to increase the TMP, although this remains significantly less than in sidestream operation. Fouling control is achieved by scour at the membrane surface, usually from aeration with the movement of bubbles close to the membrane surface generating the necessary liquid shear velocity.
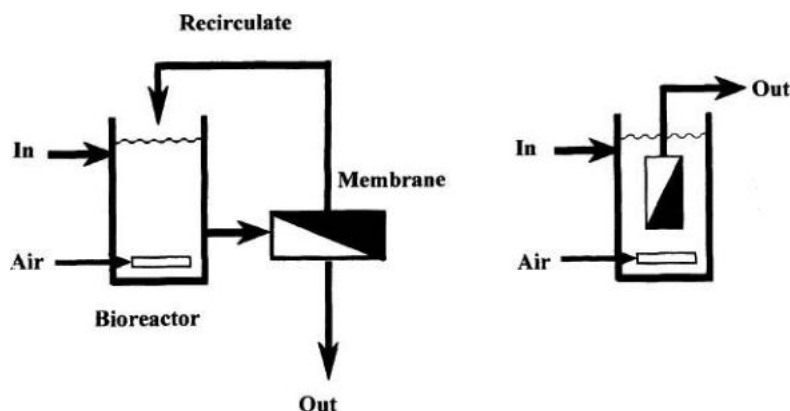
Figure 4.1: Configurations of MBRs: Sidestream (left) and submerged (right)

4.2 MEMBRANES AND BIOLOGICAL PROCESS

The coupling of a membrane to a bioreactor has attracted increasing interest both academically and commercially because of the inherent advantages the process offers over conventional biological wastewater treatment systems. Of these, process intensification and product water quality are the most obvious. The permeate from the membrane is free from solids and macro-collodial material. Typical water product qualities are <5 mg l^{-1} suspended solids and <1 NTU turbidity. Complete retention of all suspended matter is attainable, including bacteria and viruses, such that MBR effluents can be of a quality suitable for discharge to sensitive regions, further purification by dense membrane processes (such as reverse osmosis) or even recycling.

Similar benefits arise in the biological process. Sludge age and HRT are completely independent, removing some of the acknowledged operational limitations of the activated sludge process. Therefore MBRs can be operated at low HRTs and long sludge ages without washout of biomass common in activated sludge. The membrane effectively nullifies problems of filamentous

growth and degassing sludges, enabling optimal control of the reactor in terms of the residence time of the micro-organisms.

The greatest restriction of the activated sludge process is the limitation the sedimentation process places on the biomass concentration that can be maintained. The inclusion of a membrane produces an order of magnitude intensification of the process. Mixed liquor concentrations of up to $25,000$ mg^{-1} can be easily maintained during municipal wastewater treatment, while in some industrial situations this can be increased to $80,000$ mg l^{-1}. This intensification is directly translated into a reduction in reactor volume and hence footprint.

The combination of high biomass concentrations and the complete retention of solids allow the process to be operated at low organic loading rates. Reported F:M ratios as low as 0.05–0.15 d^{-1} are common. Development of specialised micro-organisms such as nitirifers promotes improved removal of nitrogen compounds and refractory organics. The lower loading rates also reduce excess sludge production down to below half that commonly encountered in activated sludge. At the limit, zero production of sludge has been achieved whereby all incoming organic feed is utilised for cell maintenance rather than growth (Müller et al., 1995).

The advantages described above are offset by several disadvantages which have to date restricted the widespread application of MBR technology, the most significant being the cost incurred largely by the membrane itself. Membrane component costs are approximately proportional to plant size as opposed to conventional plants that show a downward economy of scale. This imposes a limit to the maximum size of an economically viable MBR plant.

Operational problems also exist. Common to all membrane systems is the inevitable fouling that occurs. This limits the maximum flux obtainable and/or leads to substantial cleaning requirements. Moreover, the high biomass concentrations that are a facet of the process can result in aeration problems. The majority of the air supplied is required for cell maintenance, rather than for aerobic degradation. In the case of submerged systems aeration also acts as the source of scour at the membrane surface. The viscosity of these sludges becomes significant when the biomass concentration exceeds $25,000$ mg l^{-1}. Aeration and mixing can then become rate limiting to the process.

Though design and operation of an MBR can be considered in terms of its components parts as a biological process (Chapter 3) and a membrane filtration unit (Chapter 2), such a view fails to address aspects specific to MBRs. This added complexity occurs as the two processes work in conjunction with one another rather than as connected but independent unit operations. In particular increased shear and the absence of the clarifier are attributed to much of the differences in performance.

The fundamental differences in the biology of an MBR compared to an activated sludge process are not yet clear. A limited amount of information is also available on how descriptive variables such as the floc structure, respiration rate, species diversity and off gas production are affected by the changes in operation. Table 4.1 shows results of one study from Cicek *et al.*, (1999a) which compared the performance of an activated sludge plant with a sidestream MBR, both fed with the same synthetic sewage. The flocs in the MBR were shown to be significantly smaller and more active with a higher volatile fraction in the mixed liquor and a greater diversity of species especially in terms of free swimming bacteria. Enzyme activity was also seen to be higher in the MBR and this was attributed to washout in the activated sludge system.

Table 4.1: Performance comparison between activated sludge (AS) and MBRs (Cicek *et al.*, 1999a)

Parameter	AS	MBR
Sludge age (d)	20	30
COD removal (%)	94.5	99
DOC removal (%)	92.7	96.9
TSS removal (%)	60.9	99.9
Ammonical N removal (%)	98.9	99.2
Total P removal (%)	88.5	96.6
Sludge production (kgVSS COD^{-1} d^{-1})	0.22	0.27
Mean floc size (μm)	20	3.5

4.3 AEROBIC MBRs FOR MUNICIPAL WASTEWATER TREATMENT

The application of MBRs to domestic wastewater treatment is a major area of research at both laboratory and pilot scale with reactor volumes ranging from a few litres to >20 m^3. In the following section the performance and operation of MBRs treating municipal-type wastewater is discussed (Tables 4.2–4.4).

4.3.1 Loading rates and retention times

Reported volumetric loading rates range between 1.2 to 3.2 kg COD m^{-3} d^{-1} and 0.05 to 0.66 kg BOD_5 m^{-3} d^{-1} with corresponding removal efficiencies >90% and 97% (Table 4.2). Steady state effluent BOD concentrations have been consistently <10 mg l^{-1}, irrespective of the wide range of influent concentrations encountered, e.g. from >100 mg l^{-1} (Murakami *et al.*, 1999) to >250 mg l^{-1} (Martyn *et al.*, 1999).

Although many loading rates are similar to those for activated sludge, removal efficiencies are higher. Between 90 and 98% COD removal efficiencies have been reported, corresponding to effluent COD concentrations generally of 40 mg l^{-1} (Table 4.2). The equivalent removal efficiency in the activated sludge process operated at the same low sludge loading rates as MBRs is 75 to 85% (Dorau *et al.* 2000). Côté *et al.*, (1997) and Dorau *et al.* (2000) attributed the improved COD removal to the combination of complete particulate retention by the membrane, including suspended COD and high molecular weight organics, as well as the avoidance of biomass washout problems common in activated sludge. In particular, a secondary benefit is the provision of stable conditions for the growth of specialised micro-organisms that are able to remove slowly degradable components.

Performance appears to be relatively insensitive to HRT with values between 2 and 24 h resulting in very high removal percentages. Sludge age also appears to have little influence on effluent quality, with sludge ages between 5 and 3500 d (effectively zero sludge wastage) being used. Comparison across the reported systems shows a slight improvement in removal efficiency with increasing sludge age up to 30 d, whereafter no further improvement is shown. Effluent COD in a hollow fibre MBR has been maintained at below <16 mg l^{-1} despite a five-fold change in the sludge retention time (Côté *et al.*, 1997); substantially outperforming an activated sludge plant operating in parallel that produced a 40–50 mg l^{-1} COD effluent.

4.3.2 Nutrient removal

The complete retention of micro-organisms by the membrane can encourage the growth of specialised micro-organisms such as *Nitrosomonas* and *Nitrobacter*. Complete nitrification has been demonstrated in MBRs at sludge ages of 5–72 d and organic loading rates of 0.05–0.66 kg BOD m^{-3} d^{-1} (Table 4.2). These BOD loading rates are within ranges suitable for nitrification in comparable processes. Sludge age has been shown to have an influence on nitrification in MBRs, with reported ammonia removal efficiencies increasing from 80 to 99% on increasing the sludge age from 10 to 50 d (Côté *et al.*, 1997) and from 94 to 99% on doubling the sludge age from 5 to 10 d (Fan *et al.*, 1996). The nitrifier content of an MBR mixed liquor appears to be similar to that of an activated sludge plant, with ammonia oxidation capacities of around 0.2 mmolN $gMLSS^{-1}$ h^{-1} reported for both processes when operated at low sludge loading rates (Müller *et al.*, 1995). However, the mean nitrification activity for the MBR has also been demonstrated to be more than double that of an equivalent activated sludge plant: 2.28 g NH_4-N kg $MLSS^{-1}$ h^{-1} for the MBRs compared to 0.96 g NH_4-N kg $MLSS^{-1}$ h^{-1} for a conventional process according to Zhang *et al.*, (1997). These

authors further showed the nitrification rate to be directly related to the average floc size of the biomass, the smaller floc sizes encountered in the MBR process accounting for the improved nitrification rate observed.

Total nitrogen removal through the inclusion of an anoxic zone is common in MBR systems. Operation usually includes a separate tank (Côté *et al.*, 1997; Ghyoot *et al.*, 1999) although intermittent aeration systems have also been developed (Nah *et al.*, 2000). As with conventional biotreatment, intermittent aeration performance has been shown to be dependent on both the anoxic and oxic cycle time and the BOD/TN ratio, with total nitrogen removal dropping from >80% to c50% on decreasing the BOD/TN ratio from >2 to <1 (Nah *et al.*, 2000). Using intermittent aeration, Suwa *et al.*, (1992) achieved a denitrification rate of 0.0074 gN VSS^{-1} d^{-1} in a sidestream MBR treating synthetic wastewater. A similar operating protocol resulted in 92.6% denitrification in a submerged MBR (Chiemchaisri *et al.*, 1999). At pilot-scale, a two-stage aerobic/anoxic bioreactor coupled with an ultrafiltration membrane system has been used to treat landfill leachate (Lubbecke *et al.*, 1995). 'Complete elimination of nitrogen' was achieved at loadings of up to 4 kg NH_4-N m^{-3} d^{-1} for nitrification and 5 kg NO_3-N m^{-3} d^{-1} for denitrification. Suwa *et al.*, (1992) related denitrification rate to BOD loading such that no denitrification occurred below a loading rate of 0.438 g l^{-1} d^{-1} and thereafter increased linearly with loading rate. They type of supplementary carbon added also has an effect; MBRs giving 90% nitrogen removal can be operated at double the loading rates with methanol instead of acetic acid (Ghyoot *et al.*, 1999). The maximum loading rate for complete nitrification (0.16 kg N kg SS^{-1} d^{-1}) dropped by 50% when a positive displacement pump used for recirculating the biomass was replaced with a centrifugal pump. This demonstrated the impact of biomass shear.

Phosphorus removal in MBRs is a major area of interest as the need to reduce nutrient loads becomes more important. Reported phosphorus removals range from 11.9% (Côté *et al.*, 1997) to 75% (Ueda and Hata, 1999). Assimilation alone does not account for all phosphorus removal. For example, Dorau *et al.* (2000), observed a 40% phosphorus removal efficiency by a submerged MBR and a mass balance revealed that for assimilation to be the sole mechanism, 20% of the biomass would have to be phosphorus. Stable phosphorus removal however, has been demonstrated by metal coagulant dosing achieving a removal efficiency of 80% or more at molar ratios of 1:1 Al or Fe:P (Buisson *et al.*, 1998).

4.3.3   Biomass and sludge

Reported mixed liquor concentrations of between 10,000 and 20,000 mg l^{-1} are common for MBRs (Table 4.2), with some zero-sludge wastage processes operated at biomass concentrations up to 50,000 mg l^{-1} (Houten and Eikelboom,

1997; Müller *et al.*, 1995). Biomass separation MBRs typically produce less sludge than other comparable wastewater treatment processes owing to their operation at relatively long sludge ages and low sludge loading rates. Reported sludge production rates vary between 0 to 0.34 kg MLSS kg COD^{-1} removed (Table 4.2). However, in some cases the sludge production has been close to or greater than in conventional processes (Chaize and Huyard, 1990; Murakami *et al.*, 1999). From the reports in Table 4.2 it would appear that little or no sludge is produced at loading rates of around 0.01 kg COD kg $MLSS^{-1}$ d^{-1}.

Comparison of sludge composition has shown the ash content to differ little between an MBR and activated sludge, the former increasing from 21.6 to 23.5% over a 300 d operating period (Müller *et al.*, 1995). Trace metal analysis showed that the MBR sludge was higher in Cd, Cr and Ni; lower in Cu, K, Mg, Ag and Hg and similar in Fe, Zn, Cu, Pb, P and N. Analysis of the Fe and Al content of a submerged hollow fibre MBR revealed a slight increase in concentration during the initial 80 d operation period, and then a decline thereafter (Murakami *et al.*, 1999). A reasonable correlation between DO and metal concentration was demonstrated, indicating that a greater proportion of the metals remained dissolved at low DOs. Biomass separation membrane bioreactor sludge has also been shown to be generally more readily dewaterable according to capillary suction time (CST) measurements (Murakami *et al.*, 1999), and for dewaterability to increase with increasing sludge age (Bouhabila *et al.*, 1998). Conversely, specific resistance to filtration (SRF) measurements for sludge from both an MBR and an ASP operating in parallel have revealed the MBR sludge to have SRF values of $0.3-3 \times 10^{12}$ m kg^{-1}, three orders of magnitude greater than for the comparable activated sludge (Heiner and Bonner, 1999). The authors accounted for this in terms of the large fraction of smaller sized particles found in the MBR sludge.

4.3.4 Flux and hydrodymanics

Flux rates in MBRs range from 5 to 300 l m^{-2} h^{-1}, with specific flux values ranging from approximately 20 to 200 l m^{-2} h^{-1} bar^{-1} (Table 4.3). The exact flux at which a system operates is dependent upon a number of complex inter-related parameters, including transmembrane pressure (TMP), crossflow velocity, pore size and biomass characteristics. A standard design flux of 0.5 m^3 m^{-2} d^{-1} (20.8 l m^{-2} h^{-1}) has been suggested for a submerged plate-and-frame MBR with a membrane pore size of 0.4 μm (Ishida *et al.*, 1993); yielding a specific flux of 75–100 l m^{-2} h^{-1} bar^{-1} depending on the TMP. This compares to an instantaneous specific flux of around 125–175 l m^{-2} h^{-1} bar^{-1} for a hollow-fibre submerged microfiltration (MF) system backwashed every 15–30 min (Côté *et al.*, 1997). Sidestream MBRs, on the other hand, can be subject to significant

flux decline. The specific flux of a polymeric UF membrane operating at a TMP of 1–2 bar and crossflow velocity of 1.5 m s^{-1} decreased from 90 l m^{-2} h^{-1} bar^{-1} to 15 l m^{-2} h^{-1} bar^{-1} over 80 d of operation (Chaize and Huyard, 1991). Ceramic membrane-based MBRs are less susceptible to fouling, and have been operated at high fluxes (80 to 100 l m^{-2} h^{-1} at 1 bar TMP) for an extended period of time (15 d) without flux decay (Trouve et $al.$, 1994).

In general, sidestream systems generate higher actual flux rates than submerged systems by operating at higher pressures. The greater pressures require greater levels of liquid shear at the membrane surface to control fouling. Operating conditions have varied with pressures of 1 to 5 bar and crossflow velocities around 1 to 3 m s^{-1} (Table 4.3) but can be as high as 8.7 m s^{-1} (Krauth and Staab, 1994). Ahn et $al.$, (1998) operated a 15 kDa tubular ceramic side stream MBR at 147, 227 and 366 kPa and a crossflow velocity of 4 m s^{-1}. At pseudo steady state the flux was between 105–180 l m^{-2} h^{-1}, equating to a specific flux of 50–70 l m^{-2} h^{-1} bar^{-1}, which is somewhat lower than values obtained for submerged MBRs operated with polymeric membrane materials.

Table 4.2: Biological performance of aerobic MBRs treating domestic wastewater

Wastewater	Type*	V (m³)	HRT (h)	θ$_x$ (d)	Sludge load (kg COD kg⁻¹ d⁻¹)	Organic load (kg m⁻³ d⁻¹)	BOD^a, COD^b, NH$_3^c$, P^d (mg l⁻¹) In	Out	MLSS (kg)	Sludge yield (d⁻¹)	Air^e (m³ h⁻¹) DO^f (mg l⁻¹)	Reference
Raw domestic	HF/S/PE	3.12	13.4	72	0.021^a	0.245^a	135^a 34^c 4^d	1.3^a 7.1^c 1^d	12.9	—	18^e	Ueda and Hata, 1999
Municipal	HF/S	1	7.3	50	0.1	1.2^b	457^b 38^c 11.9^d	10.5^b 11^d 9.4^d	15	0.2^b	—	Buisson et al., 1998
Municipal	HF/S	2.6–3.9	10–16	3120	0.07^{b,v}	2.4^b	900^b	45^b	<23	0^b	—	Rosenberger et al., 1999
Municipal	T/SS/C	1	24	25	0.2^{b,v}	0.18^a 0.49^b	182^a 488^b 35^c	<10^a 20^b <1^c	2.5^v	0.26^{b,v}	>3^f	Trouve et al., 1994
Municipal	HF/S	1	7.3	50	0.1	1.2^b	457^b 38^c 11.9^d	10.5^b 11^d 9.4^d	15	0.2^b	—	Buisson et al., 1998

Table 4.2 *continued*

Wastewater	Type*	V (m³)	HRT (h)	θₓ (d)	Sludge load (kg COD kg⁻¹ d⁻¹)	Organic load (kg m⁻³ d⁻¹)	BOD[a], COD[b], NH₃[c], P[d] (mg l⁻¹) In	Out	MLSS (kg)	Sludge yield (d⁻¹)	Air[e] (m³ h⁻¹) DO[f] (mg l⁻¹)	Reference
Municipal	T/SS/C	1	24	25	0.2[b,v]	0.18[a] 0.49[b]	182[a] 488[b] 35[c]	<10[a] 20[b] <1[c]	2.5[v]	0.26[b,v]	>3[f]	Trouve *et al.*, 1994
Urban	HF/S	0.02	3.3	10	0.45[b]	3.2[b]	420[b]	42[b]	6.8	0.34[h]	1.8[e]	Bouhabila *et al.*, 1998
Urban	HF/S	0.02	3.3	20	0.39[b]	3.2[b]	450[b]	29[b]	8.3	0.2[b]	1.8[e]	
Municipal	T/SS/C	1.5	15	20	0.16[b]	0.62[b]	388[b] 40[c]	15[b] <0.5[c]	2.6[v]	0.1[b]	>4[f]	Fan *et al*, 1998
Municipal	T/SS/C	1.5	7.5	5	0.41[b]	0.55[b,v]	310[b] 31[c]	19[b] 2[c]	1.8[v]	—	>4[f]	
Municipal	HF/S	—	2	5–10	0.28[b]	2.24[a] 4.27[b]	187[a] 356[b] 28[c]	<5[a] 16[b] 5.6[c]	5–15	—	—	Côté *et al.*, 1997

Table 4.2 *continued*

Wastewater	Type*	V (m³)	HRT (h)	θ_x (d)	Sludge load (kg COD kg⁻¹ d⁻¹)	Organic load (kg m⁻³ d⁻¹)	BOD[a], COD[b], NH$_3$[c], P[d] (mg l⁻¹) In	Out	MLSS (kg)	Sludge yield (d⁻¹)	Air[e] (m³ h⁻¹) DO[f] (mg l⁻¹)	Reference
Municipal	HF/S	-	2	50	0.39[b]	2.64[a] 5.78[b]	220[a] 482[b] 39[c] 9.2[d]	<5[a] 10[b] 0.4[c] 8.1[d]	15	0.25[b]	96[c]	Côté et al., 1997
Municipal	PF/S	—	7.6–11.4	25 40	0.025–0.042[a]	0.32–0.63[a]	176[a] 79[b] 22.4[c] 3.7[d]	1.7[a] 6[b] 0.1[c] 1.2[d]	12–18	—	—	Ishida et al., 1993
Raw sewage	HF/S	21.4	13	120	0.04[a]	0.315[a]	133[a] 71[b] 32[c] 3.8	1[a] 5[b] 5.1[c] 1.1[d]	12	—	42[c]	Ueda et al., 1996, 1997
Domestic	T/SS/C	0.13	—	—	—	0.67[b]	410[b] 31[c]	54[b] 1[c]	3.7	—	—	Ghyoot et al., 1999

Table 4.2 continued

Wastewater	Type*	V (m³)	HRT (h)	θ_x (d)	Sludge load (kg COD kg⁻¹ d⁻¹)	Organic load (kg m⁻³ d⁻¹)	BOD[a], COD[b], NH₃[c], P[d] (mg l⁻¹) In	Out	MLSS (kg)	Sludge yield (d⁻¹)	Air[e] (m³ h⁻¹) DO[f] (mg l⁻¹)	Reference
Domestic +synthetic	T/SS/C	0.13	---	---	...	3[b]	7025[b] 62[c] 85[d]	187[b] 97[c] 8[d]	20.5	0.037[b,v]	...	Ghyoot et al., 1999
Domestic	HF/S	2.1	---	---	---	---	98[a] 24.7[c] 3.68[d]	0.9[a] 15[c] 2[d]	27.9	0.6	5–10[e]	Murakami et al., 1999
Domestic	HF/S	3	10.5	---	0.03[a]	0.29[a]	127[a] 44.2[b] 16[c] 3.2[d]	0.8[a] 3.9[b] 0.6[c] 2[d]	9.2	---	0.0056[e]	Futamura et al., 1994
Domestic	PF/SS	0.0045	1–8	100	0.06–0.1[b]	0.45–1.5[b]	550[b] 80[c]	<30[b] <0.5[c]	12.8	0.56[b]	...	Chaize and Huyard,1990
Domestic	T/SS	0.61	7.4–14.8	→∞	0.021[b]	0.9–2.0[b]	130[b]	<1.3[b]	40–50	0[b]	>0.6[e]	Müller et al., 1995

Table 4.2 continued

Wastewater	Type*	V (m³)	HRT (h)	θ_x (d)	Sludge load (kg COD kg⁻¹ d⁻¹)	Organic load (kg m⁻³ d⁻¹)	BOD^a, COD^b, NH₃^c, P^d (mg l⁻¹) In	Out	MLSS (kg)	Sludge yield (d⁻¹)	Air^e (m³ h⁻¹) DO^f (mg l⁻¹)	Reference
Domestic	HF/S	1.5	6–15	>3500	0.03–0.08^b	1.96^b	490^b	<15^b	<23.6		0.45^e	Chiemchasri et al., 1992
Municipal	PF/S	0.035	4.5	→∞	0.08^a	0.269^a	134^a 250^b 16.5^c	3.5^a 19^b 0.39^c	<9	0^a	1.2^e	Gander et al., 2000
Municipal	PF/S	0.035	8	→∞	0.04^a	0.115^a	134^a 250^b 16.5^c	4.1^a 16.7^b <0.1^c	<10	0^a	1.2^e	Gander et al., 2000
Domestic	T/SS	0.6	6–8	—	0.15^b 0.029^b 0.04^b	1.7–2.2^b	—	—	<50	0.19^b 0^b 0.1^b	0.5–3^f	Houten and Fikkelboom 1997
Municipal	PF/S	—	—	30–60	—	—	216^a 538^b 30^c	<5^a <24^b 0.17^d	18	0.48^a	220^e	Judd, 1997; Churchouse 1997

[71]

Table 4.2 continued

Wastewater	Type*	V (m³)	HRT (h)	θx (d)	Sludge load (kg COD kg⁻¹ d⁻¹)	Organic load (kg m⁻³ d⁻¹)	BOD[a], COD[b], NH3[c], P[d] (mg l⁻¹) In	Out	MLSS (kg)	Sludge yield (d⁻¹)	Air[e] (m³ h⁻¹) DO[f] (mg l⁻¹)	Reference
Municipal	S	—	4.98	-	0.03[a] 0.11[b]	0.36[a] 1.76[b]	115[a] 365[b] 22[c]	10[a] 2[b] <1[c]	16	-	—	Peters et al., 1999
Municipal	WC/S	—	91	—	0.02[a]	0.05[a]	172[a] 29[c]	1.8[a] 0.02[c]	2.5	—	7.2[f]	Martyn et al., 1997
Municipal	WC/S	-	10	—	0.25[a]	0.66[a]	256[a] 41.8[c]	4[a] 3.2[c]	2.5	-	7[f]	
Municipal	PF/S	15.5	4.5	45	0.03– 0.15[a]	—	200[a] 469[b] 41.6[c]	4[a] 61[b] 5[c]	10–39	0.26[a]	142[e]	Davies et al., 1998
Black water	PF/S	0.066	34.2	→∞		106[a] 323[b]	1a 15.1[b]			0[a]	74[f]	Jefferson et al., 2000
Grey water	PF/S	0.066	31.5	→∞		41[a] 120[b]	1.1[a] 9.5			0[a]	74[f]	Jefferson et al., 2000
Household	HF/S	0.04	10–15	→∞	—	—	127[a] 343[b] 21.4[c]	1[a] 11[b] 0[c]	6–14	—	1	Nah et al., 2000

Table 4.2 *continued*

Wastewater	Type*	V (m³)	HRT (h)	θx (d)	Sludge load (kg COD kg⁻¹ d⁻¹)	Organic load (kg m⁻³ d⁻¹)	BOD^a, COD^b, NH$_3$^{c}, P^d (mg l⁻¹) In	Out	MLSS (kg)	Sludge yield (d⁻¹)	Air^e (m³ h⁻¹) DO^f (mg l⁻¹)	Reference
Municipal	HF/S	1	4.8	—	0.09	2.3^b	457^b 55^{tn}	16^b 17^{tn}	26	0.2^b	0.5–1.5^f	Côté *et al*, 1997
Municipal	HF/S	1	6.5	—	0.08	1.7^h	457^b 55.1^{tn}	13^b 14^{tn}	21	—	0.5–1.5^f	
Municipal	HF/S	1	9.2	—	0.07	1.2^b	457^b 55.1^{tn}	9.9^b 11.1^{tn}	16	-?	0.5–1.5^f	

a =BOD$_5$, b = COD, c = ammonia, d = phosphorus, e= aeration rate, f = DO, g = turbidity, h = suspended solids, tn = total nitrogen, v = volatile. Type* HF = hollow fibre, PF = plate and frame, T = tubular, WC = woven cloth, S = side stream, S = submerged, SS= side stream, C = ceramic.

[73]

The influence of shear on sidestream MBR performance has been demonstrated in a number of studies. In a sidestream ceramic UF membrane MBR under constant flux conditions, the rate of pressure increase has been shown to decrease commensurately in magnitude as the wall shear rate increased from 890 s^{-1} to 32,440 s^{-1} (Tardieu et al., 1998). During laminar flow conditions, the pressure increased exponentially from 4 kPa h^{-1} to 70 kPa h^{-1}; whereas during turbulent conditions the pressure increased linearly at a rate of 0.2 kPa h^{-1}. Both the shear-induced diffusion and inertial lift models were found to be inapplicable due to the biological nature of the solids, as changes in the biological conditions of the reactor contribute to changes in the membrane resistance, in addition to the system hydrodynamics. Defrance et al. (2000) investigated the fouling of a ceramic sidestream MBR with a nominal pore size of 0.1 μm under a range of different crossflow velocities with increasing TMP. In all cases, the flux increased linearly with TMP up to a critical point, beyond which the flux levelled out. The critical point increased with increasing shear rate, i.e. crossflow velocity. Critical points of 90 l m^{-2} h^{-1} at 0.9 bar and 46 l m^{-2} h^{-1} at 0.3 bar have been reported as the crossflow velocity decreased from 4 to 2 m s^{-1}. Similar observations have been reported by Jeannot (1992) and Magara and Itoh (1991). However, the sensitivity of critical pressure to crossflow velocity has been variable, reflecting the criticality of the hydraulic resistance of the deposit and the dependency of this on the system microbiology (Tardieu et al., 1998).

A characteristic of submerged MBR systems is the development of a stable flux for long periods of time without the need for chemical cleaning because the TMP is small and flux does not exceed a critical level. Reported operating pressures for submerged systems range from 0.03 to 0.3 bar (Table 4.3) with correspondingly low actual flux rates both for flat plate and hollow fibre configurations. An example is described by Visvanathan et al., (1997) who reported a stable flux rate of 8.3 l m^{-2} h^{-1} at a TMP of 0.13 bar. Effects of crossflow velocity on submerged MBR operation has been less well explored than in the sidestream case since crossflow is generated by air sparging within the bioreactor and is not easily defined. Moreover, the action of the air sparge is configuration dependent. In plate-and-frame systems, the bubble movement creates an associated superficial liquid velocity across the rigid membrane surface (Kishino et al., 1996); whereas in hollow fibre systems, the aeration creates vibration of the individual free-moving fibres (Ueda et al., 1997; Ahn et al., 1999).

Beneficial effects of increased air flow have been demonstrated for both hollow fibre (Bouhanbila et al., 1998; Shimizu et al., 1996b; Ueda et al., 1997)

and plate-and-frame (Kishino *et al.*, 1996) systems. Shimizu *et al.*, (1996b) observed a linear relationship between air–liquid two-phase flow velocity and flux above velocities of approximately 0.3 m s^{-1}. Ueda *et al.*, (1997) showed a reduction in the required TMP, from 25 to 18 kPa, to obtain a constant flux of 15.4 l m^{-2} h^{-1} when the aeration rate was increased from 0.3 to 1.1 m^3 min^{-1}. Kishino *et al.*, (1996) calculated the superficial liquid velocity in a plate and frame system to be between 0.3 and 0.5 m s^{-1} which agrees with experimental measurement by Ueda *et al.*, (1997) at approximately 0.4 m s^{-1}.

Mukai *et al.*, (2000) showed a growth phase effect on flux decline. In a batch activated sludge trial with a UF membrane the flux dropped to 20% of its original value when the bacteria where in either the stationary or death phases of growth as opposed to only 50% during the logarithmic phase. Differences were also observed when testing different bacterial strains during their stationary phase of growth. The ratio of retained sugar to protein was postulated as one of the factors affecting the relative flux decline as a result of separate or combined interactions at the membrane surface. However, Ishiguro *et al.*, (1994) showed no significant effect on the flux by either dissolved organic carbon, sugar, protein or gel-permeation chromatography area; the latter being indicative of the molecular weight distributions of organic solutes.

4.3.5 Configurations

Few direct comparisons of the effect of configuration have been carried out, although configuration is known to effect the relationship between TMP and crossflow velocity. A submerged hollow fibre MBR had a specific flux of 50 to 65 l m^2 h^{-1} bar^{-1} compared to 115 l m^{-2} h^{-1} bar^{-1} for a flat plate configuration, all other performance determinants being similar (Günder and Krauth, 1998). A tubular sidestream MBR tested alongside the submerged systems had specific flux values of 40–60 l m^{-2} h^{-1} bar^{-1}. Le Clech *et al.*, (2000) observed a similar trend when comparing a submerged polysulphone plate-and-frame with a polyethylsulphone tubular sidestream MBR for treating domestic grey water. The two systems were operated at TMPs of 0.06 and 2.5 bar with specific flux rates of 86 and 23 l m^{-2} h^{-1} bar^{-1} respectively. It appears that a submerged MBR will have specific fluxes two to four times that of an equivalent sidestream configuration.

4.3.6 Materials and pore size

A wide range of membrane materials have been tested with pore sizes usually in the 0.02–0.5 μm size range, i.e. from mid-range UF to MF (Table 4.3),

Materials used include ceramics, for some sidestream systems, and both unmodified and surface-modified polymeric materials such as polyethylene, polypropylene and polysulphone. Comparison of a MBR configuration based on different membrane materials has shown a hydrophilic polyacrylonitrile membrane to give a flux of up to twice that of a polyolefin material (Magara and Itoh, 1991). On the other hand, some studies have shown negligible difference in performance between some polymeric materials (Thomas *et al.*, 2000).

Several studies have demonstrated the increased fouling resistance of ceramic based sidestream MBRs, with equilibrium fluxes and specific fluxes of 100–180 l m^{-2} h^{-1} and 75–120 l m^{-2} h^{-1} bar^{-1} respectively being recorded for high-shear systems by Ahn *et al.*, (1998). According to this study and others the stabilised flux is insensitive to pore size, similar values being measured at membrane pore sizes of 15 kDa, 300 kDa and 0.1 μm. On the other hand, evidence suggests that the fouling rate is greater for MF-based sidestream systems than for UF MBRs (Thomas *et al.*, 2000). In a submerged flat plate system the rate of flux decline, rather than the ultimate stable flux, was shown to be pore size dependent. A 60-fold decrease in the rate of flux decline was reported as the pore size decreased from 5 to 1 μm although the ultimate flux stabilised at 5 l m^{-2} h^{-1} in all cases (Gander *et al.*, 1999).

The benefit of hydrophilicity has been demonstrated in some studies. For example, for a submerged 0.1 μm polyethylene HF membrane-based MBR treating domestic wastewater a pressure increase of 1 m Aq yr^{-1} was noted when the membrane was hydrophobic compared with a rate of 0.5 m Aq yr^{-1} when it was hydrophilic (Futamura *et al.*, 1994). In a study of a plate and frame submerged 0.5 μm polypropylene hydrophobic membrane, no flow at all was recorded at a TMP of 0.06 bar whereas stabilised fluxes of around 10 l m^{-2} h^{-1} were obtained for a modified membrane material (Gander *et al.*, 1999). This would seem to reflect the propensity for membrane fouling by proteinaceous materials, as reported by Till *et al.* (1998).

4.3.7 Sludge concentration

Sludge concentration might be expected to have a profound influence on MBR performance owing to its effect on both the dynamic layer thickness and the viscosity. The latter affects both the sludge circulation, because of changes in hydrodynamics, and the shear stress at the filtration cake surface. The relationship between biomass concentration and flux has been studied in great detail. Logarithmic expressions are the most common (Magara and Itoh, 1991; Thomas et al., 2000) although some linear expressions against mixed liquor concentration raised to the power of −2.66 (Sato and Ishii, 1991) and −0.5 (Shimizu et al., 1996a) have been developed. Shimizu et al., (1996a) operated a submerged hollow fibre MBR with a total membrane surface area of 8 m^2 at a 30 kPa TMP and an air flow rate of 7.2 m^3 m^{-2} h^{-1}. Below 8,000 mg l^{-1} a linear relationship between flux rate and MLSS was observed; the flux then decreased by an order of magnitude as the MLSS increased to ca. 18,000 mg l^{-1}.

The exact relationship between viscosity and sludge concentration is unclear but an exponential relationship appears the most common (Manem and Sanderson 1996). Rosenberger et al., (1999) operated a sidestream tubular MBR over a range of MLSS concentrations and reported a viscosity increase from 8.5 mPa s to 75 mPa s for the suspended cells as the biomass concentration increased from 13,000 to 57,000 mg l^{-1}. Further investigation into the rheology of sludge revealed it to be pseudoplastic, with increased shear stress leading to a decrease in viscosity. Nagaoka (1999) noted that intermittent aeration in a MBR enhanced extracellular polymeric substance degradation which also reduced viscosity. In sidestream operation the increase in viscosity can be particularly important because of the increased frictional losses during pumping. Manem and Sanderson (1996) reported that a crossflow velocity of 6 m s^{-1} was required to maintain turbulent flow in a 4 mm tube when the viscosity was 6 cp, which equated to a biomass concentration of 20,000 mg l^{-1}. The relative pressure drop at this viscosity increased by more than an order of magnitude as the cross flow velocity was increased from 1 m s^{-1} to 6 m s^{-1}. At the lower effective crossflow velocities prevailing in submerged systems, much higher biomass levels are reported without problems e.g. 25,000 to 30,000 mg l^{-1} (Rosenwinkel, 1997) and 35,000 mg l^{-1} (van Dijk and Roncken, 1997).

4.3.8 Energy consumption

Membrane bioreactor power requirements came from pumping feed water, recycling retentate, permeate suction (occasionally) and aeration (Owen et al.,

1995). As already stated, submerged MBRs generate the necessary crossflow velocity through air movement in the reactor and the necessary pressure through the hydraulic head above the membrane, with additional permeate suction in some cases. In contrast, sidestream systems have large pumping requirements to circulate biomass around the membrane loop at sufficiently high pressures and velocities. Consequently, the overall energy requirements for submerged systems tend to be lower than for sidestream operation (Table 4.3).

Côté et al., (1998) reported energy consumption rates of 2–10 kWh m^{-3} for sidestream operation and 0.2–0.4 kWh m^{-3} for submerged operation. Côté et al., (1997) recorded an energy consumption of 0.3 kWh m^{-3} in a submerged hollow fibre MBR, of which 0.28 kWh m^{-3} was required for aeration. A recent review of energy consumption data (Gander et al., 2000a), based on the assumption of centrifugal pumps being used for both biomass circulation and aeration at an overall pumping efficiency of 60%, revealed very substantial different power requirements between the submerged and sidestream configurations. In the sidestream mode, aeration accounted for between 20–50% of the total power requirements, whereas it accounted for between 80 and 100% of the total in submerged systems. The overall power demand for submerged MBRs was shown to be up to three orders of magnitude less than for a comparable sidestream system.

4.3.9 Disinfection

It has been noted that MBRs are capable of removing high numbers of bacteria and viruses (Table 4.3). While pore sizes may be greater than the diameter of the organisms, removal has been greater than might have been expected. Chiemchaisri et al. (1992a) observed an improvement in virus removal as a gel layer was formed on the membrane surface. In contrast, Cote et al. (1997) attributed high phage removal to high MLSS concentrations in the bioreactor, phage adsorbing onto the solids. Studies of bacteria removal in physical separation appear to confirm the view that formation of a gel layer decreases the effective cut-off (Till et al., 1998). However, relatively large pore sizes (5 μm) gave 3–5 log rejections compared with 8 log rejections of total coliforms with smaller pores (0.4 μm) in similarly configured systems (Gander et al., 1999). Ueda and Horan (2000) used the same type of membranes (0.4 μm pore size) in a bench-scale MBR to determine the relative contributions of the membrane and the biomass in phage and bacteria removal. They observed that in the absence of mixed liquor biomass phage removal was negligible—the phage having a mean size of 0.2 μm, half that of the membrane pore size. As membrane filtration resistance increased, so did phage removal, indicating that membrane fouling played a significant role in removal through a reduction in effective pore size.

Jefferson *et al.*, (2000) noted that disinfection levels achievable by MBRs during greywater treatment were good enough to require no further treatment of the recycled water before non-potable use.

4.4   AEROBIC MEMBRANE BIOREACTORS TREATING INDUSTRIAL WASTEWATERS

Since the early 1980s MBR technology has been successfully applied to a wide range of industrial wastes. Applications include oily wastes (Knoblock *et al.*, 1994; Seo *et al.*, 1997), food wastes (Krauth and Staab, 1993; Mallon *et al.*, 1999), tannery effluents (Yamamoto and Win, 1991; van Dijk and Roncken, 1997) and landfill leachates (Pirbazari *et al.*, 1996; Mishra *et al.*, 1996).

4.4.1 Operating conditions of the bioreactor

Reported organic loading rates range between 0.25–16 kgCOD m^{-3} d^{-1} with corresponding removal efficiencies of 90–99.8% (Table 4.5). Loading rates are higher than municipal applications owing to the high strength of wastes. Feed concentrations of 68,000 mg l^{-1} COD (Kempen *et al.*, 1997) for a brewery effluent and 29,430 mg l^{-1} COD for an oily waste (Zaloum *et al.*, 1994) are typical examples.

The exact operating conditions of an MBR are usually waste specific. Krauth and Staab (1994) reported on the operation of a sidestream tubular MBR for the treatment of a range of different industrial wastes. A reduction in COD from 42,662 to 70.8 mg l^{-1} was achieved at a loading rate of 8.3 kgCOD m^{-3} d^{-1} for a food processing waste. Similar performance was achieved with fruit juice, cotton mill and tannery effluent at respective loading rates of 5.9, 0.25 and 3.5 kg COD m^{-3} d^{-1}. Reported sludge ages of 6–300 d were similar to those for MBRs treating municipal wastewaters (Table 4.5). However, HRTs were generally much greater in industrial applications; days rather than hours being reported (Table 4.5).

Table 4.3: Membrane performance of aerobic MBRs treating domestic wastewater

Wastewater	Type*	V (m³)	HRT (h)	Pore size (μm)	x-flow (m s⁻²)	Pressure (bar)	Initial flux (l m⁻² h⁻¹)	Final/mean flux (l m⁻² h⁻¹)	Specific flux (l m⁻² h⁻¹ bar⁻¹)	Area (m²)	Energy (kWhm⁻¹)	Reference
Raw domestic	HF/S	3.12	13.4	0.4	—	4 kPa	16.6	—	4+5	20	2.4	Ueda and Hata, 1999
Municipal	HF/S	3.9	15.6	0.2	—	0.04 0.15–0.3	18	60–120	<1ʰ	13.9	—	Doreau et al., 2000
Municipal	PF/S	—	7.6–11.4	0.4	0.5	30 kPa 0.3	20.8	69.4	<1ʰ	...	...	Ishida et al., 1993
Municipal	HF/S	—	2	0.02	—	0.2	35	175	0.14ᵍ	—	0.3	Côté et al., 1997
Municipal	HF/S	—	2	0.02	—	0.2	25	125	0.24ᵍ	8mods	0.3	
Raw sewage	HF/S	21.4	13–16	0.1	0.4	0.15	12.1	80.5	Ndʰ	4	2	Ueda et al., 1996, 1997
Domestic	HF/HS	0.45	12	0.1	0.3–0.5	0.3	16.6	55	—	8	—	Shimizu et al., 1996
Domestic	T/SS/C	0.13	-	0.1	4.7	-	125	-	-	0.05	.	Ghyoot et al., 1999

Table 4.3 *continued*

Wastewater	Type*	V (m³)	HRT (h)	Pore size (μm)	x-flow (m s⁻²)	Pressure (bar)	Initial flux (l m⁻² h⁻¹)	Final/mean flux (l m⁻² h⁻¹)	Specific flux (l m⁻² h⁻¹ bar⁻¹)	Area (m²)	Energy (kWhm⁻¹)	Reference
Domestic	T/SS/C	0.13	—	0.1	4.7	—	38	250	—	0.05	—	Ghyoot et al., 1999
Domestic	T/SS	2.75	2.3	0.025	8.7	3	300	100	—	3.96	14·29	Krauth and Staab, 1994
Municipal	T/SS/C	1	24	0.1	1.5–3.5	1	60–80	60–80	—	1.1	—	Trouve et al., 1994
Domestic	HF/S	2.4	—	0.4	—	—	16.6	—	—	16.8	—	Murakami et al., 1999
Domestic	HF/S	3	—	0.1	—	1 mAq / 9 mAq	6.6 / 25	—	—	7.34	—	
Grey water	PF/S	0.07	—	0.4	0.5	0.03	7.9	263	<2ᵍ	0.24	4	Jefferson et al., 2000
Municipal	PF/S	0.035	—	0.4	—	0.03	7	233	<0.15ᵍ	0.24	—	Gander et al., 2000
Municipal	PF/S	0.035	—	5	—	0.03	5	166	<0.4ᵍ	0.24	—	Gander et al., 2000;

[81]

Table 4.3 *continued*

Wastewater	Type*	V (m³)	HRT (h)	Pore size (μm)	x-flow (m s⁻²)	Pressure (bar)	Initial flux (l m⁻² h⁻¹)	Final/mean flux (l m⁻² h⁻¹)	Specific flux (l m⁻² h⁻¹ bar⁻¹)	Area (m²)	Energy (kWh m⁻³)	Reference
Municipal	PF/S	—	—	0.4	0.5	—	—	—	<0.4§	1500 0	—	Gander et al., 2000b;
Municipal	PF/S	—	—	0.4	—	0.1	27–32	270–320	0.55§	240	—	Judd, 1997; Churchouse 1997
Municipal	PF	—	—	0.09	1.8	0.14	25	178.5	—	25	—	Arika et al., 1977
Municipal		—	—	UF	2.5	—	120	—	—	—	—	Roullet, 1989
Municipal	T/SS	—	—	0.5	2.2	2.25	20	8.8	—	—	140	Bailey et al., 1994;
Municipal	PF/SS	—	—	0.02	2	0.212–0.312	22.9–68	—	—	2.17	—	Sato and Ishii, 1991
Municipal	S	0.275	10 (91)	12	—	0.15–0.5	26.5 (2.6)	—	—	—	—	Martyn et al., 1999
Municipal	HF/S	0.28	12	0.2	—	0.25	25	100	0.5§	0.65	—	Wilkes et al., 1999
Municipal		—	4.98	0.2	—	0.2	40	200	—	40	1	Peters et al., 1999

[82]

Table 4.3 *continued*

Wastewater	Type*	V (m³)	HRT (h)	Pore size (μm)	x-flow (m s⁻²)	Pressure (bar)	Initial flux (l m⁻² h⁻¹)	Final/mean flux (l m⁻² h⁻¹)	Specific flux (l m⁻² h⁻¹ bar⁻¹)	Area (m²)	Energy (kWhm⁻³)	Reference
Raw domestic	PF/S	3.12	13.4	0.4	—	0.17	16.6	98	0.03^b	20	2.4	Ueda and Hata, 1999
Domestic	HF/S	0.08	3–12	0.1	.	0.13	8	61.5	...	2	0.14	Viscanathan *et al.*, 1997;
Household	HF/S	0.04	10–15	0.1	.	0.04–0.06	10	166–250	—	0.5	—	Nah *et al.*, 2000
Reject liquor	T/SS/C	0.132	45.6	0.1	3	0.7 2	73 154	104 77	—	0.05	--	Ghyoot *et al.*, 1999
Domestic	HF/S	--	--	0.1	—	0.65	100	153	0.15^g	20	—	Müller *et al.*, 1995
Domestic	T/SS	0.61	7.4–14.8	0.2	1–2	5	170	34	—	2.6	0.69	

a =BOD, b = COD, c = ammonia, d = phosphorus, e= aeration rate, f = DO, g = turbidity, h = suspended solids, m = total nitrogen, v = volatile. Type* HF = hollow fibre, PF = plate and frame, T = tubular, WC = woven cloth, S = submerged, SS= side stream, C = ceramic.

[83]

Table 4.4: Disinfection by aerobic MBRs

Reference	System	Pore Size	Parameter*	Influent	Effluent	Removal (log)
Chiemchaisri (1992a, b)	sub hollow fibre	µm $0.3\ \mu m$	coliphage coliphage	$7.1\text{-}7.5$ log $6.8\text{-}7.7$ log	$1.2\text{-}3.5$ log $0.8\text{-}3.1$ log	4.6 4.6
Cote et al. (1997)	sub hollow fibre	200,000 Daltons	total coliform bacteriophage	$5.6\text{-}5.9 \times 10^7$ $1.4\text{-}3.7 \times 10^3$	$20\text{-}43$ —	$6.1\text{-}6.4$ $>3.8\text{-}>4.5$
Buisson et al., (1998)	sub hollow fibre	200,000 Daltons	faecal coliform	n/a	n/a	6.2
Jefferson et al., (2000)	sub flat sheet	$0.4\ \mu m$	total coliform	$2.5\text{-}7.4$	$1\text{-}15$	ca. 7
Churchouse and Wildgoose (1999)	sub flat sheet	$0.4\ \mu m$	faecal coliform faecal steptococcus coliphage	$0.9\ 64 \times 10^6$ $<30 \times 10^6$ $<29\text{-}6320$	<20 <11 <0.19	>5.7 $>99.9993\%$ $>99.98\%$
Gander et al. (1999)	sub flat sheet sub flat sheet sidestream tubular	$0.4\ \mu m$ $5\ \mu m$ $5\ \mu m$	total coliforms	n/a	n/a	ca. 8 ~5 5
Ueda and Horan (2000)	sub flat sheet	$0.4\ \mu m$	phage faecal coliform faecal steptococci	$1.5 \times 10^3\text{-}1.1 \times 10^8$ $8.8 \times 10^6\text{-}1.2 \times 10^7$ $5.2 \times 10^5\text{-}7.7 \times 10^3$	n/a	$2.3\text{-}5.9$ 6.86 75.83

* Coliforms and Steptococci in CFU/100 ml; phage in PFU/ml

Trials on the effects of HRT and sludge age have shown no benefits in operating at longer HRTs for treatment of an oily wastewater (Sutton *et al.*, 1994). Hydraulic retention time was varied between 1.87 and 3.74 d and the sludge age between 50 and 100 d, resulting in a COD removal efficiency >90% in most cases. Similar results have been reported for tannery wastes (Yamamoto and Win, 1991) and paper and pulp wastewater (Dufresne *et al.*, 1998). In contrast, Seo *et al.*, (1997) showed an increase in COD removal efficiency from 89 to 97% as the sludge age and HRT increased from 5 to 30 d in a batch MBR trial.

The effects of sludge age and loading rate on the nitrification capacity of a sequencing batch submerged MBR treating a tannery effluent have been described by Yamamoto and Win (1991). This was operated at sludge ages of 10, 20 and 550 d and volumetric loading rates of 3, 5 and 10 kg COD m^{-3} d^{-1}; COD removal efficiency remained >93% in all cases. However, nitrification improved as the loading rate increased with average ammonia removal efficiencies of 61, 70.6 and 86.3% respectively. In cases with very high strength wastes, initial loading rates have had to be reduced to avoid inhibition of the nitrifiers.

An important consideration in industrial effluent treatment is the removal of specific components. For example, significant degradation (>95%) of fats, oils and greases (FOG) has been shown at several sites (Sutton *et al.*, 1994), even during hydraulic shock loads (Zaloum *et al.*, 1994). This has been explained in terms of the membrane's ability to retain the large fat and grease molecules. Effluent quality was poorer, however, at one site once the influent total FOG concentration was >1,000 mg l^{-1} owing to the generation of extractable non hydrocarbon materials within the reactor (Knoblock *et al.*, 1994).

4.4.2 Biomass

Biomass concentrations between 2,000 and 40,000 mg l^{-1} have been reported with a significant number of MBRs operating at >20,000 mg l^{-1} (Table 4.4). Overall sludge yields are similar to municipal wastewaters, typically between 0.05 and 0.35 kg SS kg COD^{-1} d^{-1}. The yield will be composition dependent, e.g. similar yields have been reported for fruit juice and tannery effluents in a MBR operated at sludge ages of 6.2 and 30.8 d respectively (Krauth and Staab, 1994). A food processing wastewater had a 35% lower sludge production rate than for the tannery wastewater, even though it was operated at half the sludge age. Wastes with a higher Freon extractable hydrocarbon FOG to total FOG ratio, such as those with a high fraction of petroleum based metal working fluids, had higher net yields coefficients (Sutton *et al.*, 1994).

4.4.3   Flux and configuration

Most reports on industrial wastewater treatment in MBRs have used sidestream systems with a tubular module and a pore size between 0.001 and 0.1 μm (Table 4.6). However, submerged systems have been used for the treatment of food wastes (Mallon *et al.*, 1999) and landfill leachates (Ahn *et al.*, 1999). Consequently, in most schemes the membranes are operated at relatively high TMPs (1.5–3 bar) and crossflow velocities (1.6–4.5 m s^{-1}). This results in high actual flux rates (up to 150 l m^{-2} h^{-1}) but low specific flux rates (<100 l m^{-2} h^{-1} bar^{-1}). Like most sidestream operations, membrane fouling is inevitable. Seo *et al.*, (1997) reported a 70% flux reduction in 7 d when operating a hollow fibre system to treat an oily wastewater. Cleaning restored the flux to ca. 80% of its original value but then decayed, reaching a minimum of 2 l m^{-2} h^{-1} when operating at a TMP of 1 kg cm^{-2}. Similar results have been reported in a submerged system treating a pharmaceutical effluent over a 20–80 kPa range of suction pressures (Benitez *et al.*, 1995). However, Yamamoto and Win (1991) showed stable fluxes in a submerged hollow fibre MBR treating a tannery effluent. Performance was biomass-concentration dependent: a 3-fold increase in TMP was required as the MLSS increased from 10,900 to 18,200 mg l^{-1}.

Energy consumption in industrial plants is a major consideration. Krauth and Staab (1993; 1994) showed that specific energy consumption could be reduced by either increasing crossflow velocity or membrane area in sidestream systems. Wagner and Robinson (1999) reported on a sidestream MBR where the crossflow velocity at low overall fluxes was generated by an airlift pump. Specific energy requirements of 0.5–1.5 kWh m^{-3} were recorded compared with 1.5–3.5 kWh m^{-3} for a standard pump system.

Table 4.5: Biological performance of aerobic MBRs treating industrial wastewater

Wastewater	Type	V (m^3)	HRT (h)	θ_x (d)	Sludge load (kg COD kg^{-1} d^{-1})	Organic load (kg m^{-3} d^{-1})	BOD[a], COD[b], NH$_3$[c], P[d] In	Out	MLSS (kg m^{-3})	Sludge yield (d^{-1})	Air[e] (m^3 h^{-1}) DO (mg l^{-1})	Reference
Food	T/SS	2.75	139.2	15.9	0.5[a]	5.4[a]	31000[a] 42660[b] 1221[c] 197.5[d]	nd[a] 70.8[b] nd[c] 10.2[d]	10.9	0.094[b]	6.3[f]	Krauth and Staab, 1993
Fruit juice	T/SS	2.75	—	6.2	0.581[b]	5.98[b]	2251[b]	24.23[b]	10.3	0.335[b]	3[f]	Krauth and Staab, 1994
Vegetable	T/SS	2.75	122	15.9	0.765[b]	8.33[b]	42662[b]	70.8[b]	10.9	0.094[b]	6.3[f]	
Tannery	T/SS	2.75	—	30.8	0.231[b]	3.74[b]	7.644[b]	190[b]	16.2	0.274[b]	1.5[f]	
Cotton mill	T/SS	2.75	263	160.1	0.131[b]	0.253[h]	2778[b]	119[b]	1.95	0.048[b]	5.5[f]	
Textile	T/SS	—	—	250	—	—	6000[b]	625[b]	—	0.07[b]		Krauth and Staab, 1994

Table 4.5 *continued*

Wastewater	Type	V (m³)	HRT (h)	θ_x (d)	Sludge load (kg COD kg⁻¹ d⁻¹)	Organic load (kg m⁻³ d⁻¹)	BOD[a], COD[b], NH₃[c], P[d] In	Out	MLSS (kg m⁻³)	Sludge yield (d⁻¹)	Air[e] (m³ h⁻¹) DO (mg l⁻¹)	Reference
Dairy	T/SS	—	—	>100	—	—	2000[b]	20[b]	—	0.05[b]	—	Wagner *et al.*, 1999
Brewery	T/SS	1	102	—	0.42[b]	16[b]	54400[a] 68000[b] 13[c] 190[d]	<2[a] 1350[b] <1[c]	38	<0.1	1.5–2[f]	Kempen *et al.*, 1997
Food	HF/S	4	389		0.11[a]	3.2[a]	1853[a] 3181[b] 20.7[c] 19.2[d]	<10[a] 254[b] 0.86[c] 0.23[d]	<28	—	>0.4[f]	Mallon *et al.*, 1999
Oily	T/SS	1.9	144–240	50–75	1.36 2.72[b]	2.45–4.91[b]	5105[a] 29430[b]	<20[a] <2943[b]	1.8[y]	1.3–2%	34,000	Zaloum *et al.*, 1994

Table 4.5 *continued*

Wastewater	Type	V (m³)	HRT (h)	θ_x (d)	Sludge load (kg COD kg⁻¹ d⁻¹)	Organic load (kg m⁻³ d⁻¹)	BOD^a, COD^b, NH_3^c, P^d		MLSS (kg m⁻³)	Sludge yield (d⁻¹)	Air (m³ h⁻¹) DO (mg l⁻¹)	Reference
							In	Out				
Oily	T/SS	1.325	69.6	65	0.13^{b,v}	0.39^a 3.84^b	1147^a 11133^b	15^a 1043^b	28.9^v	0.126^{b,v}	1.3–6.7	Sutton et al., 1994; Knoblock et al., 1994
Oily	T/SS	1.325	72	36	0.21^{b,v}	0.57^a 5.54^b	1711^a 16609^b	17^a 1190^b	26.2^v	0.141^{b,v}	1.3–6.7	
Oily	T/SS	287	54	31	0.012^b	6.3^b	16609^b 14180^b	<41^a 799^b	28.7^v	0.16^{b,v}	—	Mishhra et al., 1996
Oily	HF/SS	—	88	300	0.18^b	0.36^b	1333^b 4.5^c	40.8^b 4.9^c	2	0.348^b		Seo et al., 1997
Oily	T/SS	3.78	89.7	50	0.29^{b,v}	0.25^a 1.16^b	919^a 4345^h	3^a 183^b	4.03^v	0.074^{b,v}	0.3–7.5	Knoblock et al., 1994; Sutton et al., 1994

Table 4.5 continued

Wastewater	Type	V (m³)	HRT (h)	θ_x (d)	Sludge load (kg COD kg⁻¹ d⁻¹)	Organic load (kg m⁻³ d⁻¹)	BOD[a], COD[b], NH3, p[d] In	Out	MLSS (kg m⁻³)	Sludge yield (d⁻¹)	Air[c] (m³ h⁻¹) DO (mg l⁻¹)	Reference
Oily	T/SS	3.78	44.8	50	0.2[b,v]	0.61[a] 2.97[b]	1145[a] 5543[b]	7[a] 540[b]	14.95[v]	0.112[b,v]	0.3–7.5	Knoblock et al., 1994; Sutton et al., 1994
Oily	T/SS	3.78	44.8	50	0.57[b,v]	0.64[a] 3.68[b]	1206[a] 6864[b]	34[a] 664[b]	6.5[v]	0.039[b,v]	—	
Oily	T/SS	3.78	47.2	74	0.036[b,v]	0.07[a] 0.71[b]	134[a] 1406[b]	6[a] 249[b]	19.6[v]	—	0.3–7.5	
Tannery	HF/S	0.0025	24	10	0.42[b,v] 0.46[b,v]	3[b] max10	2960[b] 9580 105[c]	161[b] 361 43.5[c]	9.5 26	0.62[v]	—	Yamamoto and Win 1991
Tannery	HF/S	0.0025	24	550	0.26[b,v] 0.28[b,v]	3[b] max 10	2960[b] 9580 105[c]	186[b] 357 45.3[c]	14.2 40	—	—	Yamamoto and Win 1991

Table 4.5 continued

Wastewater	Type	V (m³)	HRT (h)	θ_x (d)	Sludge load (kg COD kg⁻¹ d⁻¹)	Organic load (kg m⁻³ d⁻¹)	BOD[a], COD[b], NH3[c], P[d] In	Out	MLSS (kg m⁻³)	Sludge yield (d⁻¹)	Air[e] (m³ h⁻¹) DO (mg l⁻¹)	Reference
Chemical industry	—	200	48	—	—	3	4000[a] 10000[b]	<10[a] <400[v]	—	—	—	Van Dijk and Roncken, 1997
Leachate	—	180	28.8	—	—	0.7[c]	800[a] 4000[b]	<20[a] <400[b]	—	—	—	
Tannery	—	700	24	—	—	0.7[c]	3800[a] 4200[b]	<20[a] <400[b]	—	—	—	
Leachate	HF/S	9.5	240	30	0.05[c]	—	8000[a] 1100[c]	30- 300[c]	4	—	—	Ahn et al., 1999
Ferm.	RD/SS	0.03	64	600	—	0.3-0.7[TOC]	1360[c]	0.2- 3.4[c]	15.2	—	2-3.5[f]	Lu et al., 1999
Paper	HF/S	0.09	24	15	—	—	4000[a] 12000[b]	160[a] 2400[b]	24.2	—	3.7[e]	Dufresne et al., 1998

[91]

Table 4.5 continued

Wastewater	Type	V (m³)	HRT (h)	θ$_x$ (d)	Sludge load (kg COD kg⁻¹ d⁻¹)	Organic load (kg m⁻³ d⁻¹)	BOD[a], COD[b], NH₃[c], P[d]	MLSS (kg m⁻³)	Sludge yield (d⁻¹)	Air[e] (m³ h⁻¹) DO (mg l⁻¹)	Reference
Paper	HF/S	0.09	36	15	—	—	4000[a] 12000[b] 520[a] 3840[b]	14.2	—	3.7[f]	Dufresne et al., 1998
Paper	HF/S	0.09	48	15	—	—	4000[a] 12000[b] 160[a] 2160[b]	13	—	3.7[f]	
Chemical	T/SS	1	14	—	0.45[b]	9[b]	52000[b] 8[d] 6000[e] <1[d]	20	<0.05	1.5–2[f]	Kempen et al., 1997
Pharm.	T/SS	1	163	—	0.125[b]	2.5[c,m]	17000[b] 600[c] 300[b] 0[c]	20	<0.1	1.5–2[f]	

a =BOD₅, b = COD, c = ammonia, d = phosphorus, e= aeration rate, f = DO, g = turbidity, h = suspended solids, tm = total nitrogen, v = volatile. Type* HF = hollow fibre, PF = plate and frame, T = tubular, WC = woven cloth, S = submerged, SS= side stream, C = ceramic.

Table 4.6: Membrane performance of aerobic mbrs treating industrial wastewater

Wastewater	Type	V (m³)	HRT (h)	Pore size (μm)	x-flow (m s⁻²)	Pressure (bar)	Initial flux (l m⁻² h⁻¹)	Final/ mean flux (l m⁻² h⁻¹)	Specific flux (l m⁻² h⁻¹ bar⁻¹)	Area (m²)	Energy (kWhm⁻¹)	Reference
Oily	T/SS/	287	45.6	0.03	—	—	90	63.6	—	195	:	Sutton et al., 1994
Oily	T/SS/	1.9	144–240	0.03	—	2.44	70	37	15	—	—	Zaloum et al., 1994
Dairy	T/SS/	0.01	24	0.2	:	0.2	—	40	200	0.06	—	Scott et al., 1996
Food	T/SS	2.75	139	0.04	2.5	2.75	—	66	24	0.22	8–27	Krauth and Staab, 1993
Tannery	HF/S	0.0025	24	0.1	—	0.51 0.77 0.96	3.9 5.4 6.5	3.9 5.3 5.6	7.6 6.8 5.8	0.27	—	Yamamoto and Win, 1991
Industrial	T/SS	—	—	—	1.6	4	—	37	9.25	—	—	Ross et al., 1992

Table 4.6 continued

Wastewater	Type	V (m³)	HRT (h)	Pore size (µm)	x-flow (m s⁻²)	Pressure (bar)	Initial flux (l m⁻² h⁻¹)	Final/mean flux (l m⁻² h⁻¹)	Specific flux (l m⁻² h⁻¹ bar⁻¹)	Area (m²)	Energy (kWhm⁻¹)	Reference
Oily	HF/SS	—	88	0.08	—	1 kg/cm²	10.2	2	—	0.086 5	—	Seo et al., 1997
Brewery	T/SS	1	—	UF	4.5	3	—	>120	>40	0.5–3	—	Kempen et al., 1997
Chemical	T/SS	1	—	UF	4.5	3	—	>120	>40	0.5–3	—	
Pharmaceutical	T/SS	1	—	UF	4.5	3	—	>150	>50	0.5–3	—	
Landfill	T/SS	174	—	UF	4.5	3	—	>110	>36.6	5.7	—	
Industrial	PF	—	—	—	2	1.5	—	30	20	—	—	Anderson et al., 1986a, b
Pharm.	HF/S	0.02	10.5	0.001	—	0.2	72	18	90	0.3	—	Benitez et al., 1995

a = BOD, b = COD, c = Ammonia, d = Phosphorus, e = aeration rate, f = DO, g = turbidity, h = suspended solids, m = total nitrogen, v = volatile. Type* HF = hollow fibre, PF = plate and frame, T = tubular, WC = woven cloth, SS = side stream, S = submerged, C = ceramic.

4.4   ANAEROBIC MEMBRANE BIOREACTORS
TREATING WASTEWATERS

Conventional anaerobic digesters are single pass reactors with no selective solids recycle; hence the biomass and HRTs are equal. This limits the organic loading rates and operating biomass concentrations (Pillay *et al.*, 1994). One way to overcome these problems is to include membrane to enable independent control of hydraulic and solids retention. Applications have focussed on high strength wastewaters such as wine distillery effluents (Ross *et al.*, 1990), palm oil mill effluent (Fakhru'l-Razi and Noor, 1999) and Dairy (Li *et al.*, 1985), although domestic wastewater has also been treated (Wen *et al.*, 1999).

The alternatives to MBRs such as anaerobic filters and upflow sludge blanket reactors have an operational limit of around 10 kg COD m^{-3} d^{-1} for the reliable production of a high-quality effluent free from solids (Ross *et al.*, 1990). Experience with a wine distillery waste showed an increase in loading rate from 4 to 11 kgCOD m^{-3} d^{-1} by the inclusion of an ultrafiltration membrane for solids separation (Ross *et al.*, 1990). Similarly, increased removal of grease from 37 to 99% and total oxygen demand (TOD) from 45 to 90% for a wool scouring waste (Hogetsu *et al.*, 1992) and a 13% improvement in adsorbable organic halogen (AOX) removal for a kraft bleach waste (Hall *et al.*, 1995) have been reported. Biomass concentrations have also increased with levels up to 50,000 mg VSS l^{-1} being reported (Ince *et al.*, 1995). In one instance, CO_2 degassing by the membrane has caused struvite formation within the MBR resulting in an increase in solids from 15,000 to 100,000 mg l^{-1} (Brockmann and Seyfried, 1996).

4.4.1 Loading Rates

In laboratory investigations, Anderson *et al.*, (1986a, b) reported a maximum loading rate of 54 kg COD m^{-3} d^{-1} for acidogenic bacteria and 12.2 kg COD m^{-3} d^{-1} for methanogenic bacteria. In pilot studies treating brewery waste, Ince *et al.*, (1998) found that 97% COD removal efficiency was obtainable up to a maximum loading rate of 28.5 kg COD m^{-3} d^{-1}. Performance profiles across the plant demonstrated that the concentration of soluble COD was consistently two to three times higher in the reactor than observed in the permeate. Similar findings by Harada *et al.*, (1994) with a synthetic waste revealed that although significant increases in protein and sugar levels in the biomass were observed with increasing loading rate, no increase in permeate concentration could be detected. Gel permeation chromatography revealed that organics held within the biomass had molecular weights (MW) in the order of 10^6, whereas organics

found in the permeate were of the order 1500 MW. The cut off of the membrane was 10^6 MW indicating the formation of a gel layer on the membrane surface.

Removal efficiencies of >90% have been reported at a loading rate of 15 kg COD m^{-3} d^{-1} for a range of wastewaters treated with an anaerobic MBR (Li et al., 1985; Kayawake et al., 1991; Strohwald and Ross, 1992), even during organic loading rate fluctuations (Wen et al., 1999). Removal has also been shown to be reasonably stable over a wide range of loading rates. For example, Cadi et al., (1994) observed a decrease in removal efficiency of only 7% as the loading rate increased from 7.7 to 24.2 kg COD m^{-3} d^{-1}. Organic removal at mesophillic temperatures (37 °C) has been shown to be slightly better than at thermophilic ones (53 °C), although volatile fatty acids (VFA) accumulated more slower at the higher temperature indicating greater biodegrability (Hogetsu et al., 1992). In most of the reported schemes no accumulation of VFA has been reported indicating that loading rate could still be further increased. However, in one case digester overload was noted by a rapid increase in volatile acid/alkalinity ratio (0.8) during start up of the treatment of a brewery effluent. This was solved by nutrient balancing with urea (Strohwald and Ross, 1992).

The combination of the slow growth rates of anaerobic bacteria and the high influent strength of the wastes encountered means that HRTs are generally long (Table 4.7). Typical examples are 67 h for a palm oil mill effluent (Fakhru'l-Razi and Noor, 1999), 124 h for a maize processing water (Ross et al., 1992) and up to 170 h for a dairy waste (Li et al., 1985). Cadi et al., (1994) reported on the effects of HRT for the treatment of a starch based synthetic waste at a loading rate of 2 kg COD m^{-3} d^{-1} and a sludge age of 45 d. Performance deterioration occurred only once the HRT was reduced to 6 h at which point the COD removal efficiency was reduced by 12% to 78%.

4.4.2 Gas Production

The production of methane as an end product provides an additional benefit, about 22 to 26 MJ m^{-3} depending on the carbon dioxide content of the biogass. Reported methane yields range between 0.17 to 0.29 $m^3 CH_4$.kg COD^{-1} but the actual yield is heavily dependent on the wastewater source and the operating conditions (Table 4.7). The theoretical methane yield is 0.35 m^3 kg COD^{-1} (Li et al., 1985) with proportions of 65 to 75% being considered indicative of good performance (Strohwald and Ross, 1992). Biogas yields during the treatment of a brewery waste as low as 20-47% of the theoretical have been reported due to CO_2 stripping through the membrane increasing the pH within the reactor (Strohwald and Ross, 1992).

Biogass production has been shown to decline with increasing loading rate (Fakhru'l-Razi and Noor, 1999), increasing HRT (Cadi et al., 1994) and

decreasing temperature (Hogetsu *et al.*, 1992). For example, in the treatment of a brewery waste with organic loading rates as high as 28 kg COD m^{-3} d^{-1} a decrease in methane yield from 80 to 65% was observed (Ince *et al.*, 1998). Similar finding by Fakhru'l-Razi and Noor (1999) attributed the loss of gas production to the more favourable conditions of the faster growing acidogenic bacteria at higher loading rates compared to methanogenic bacteria. Kitamura *et al.* (1994) showed that maximum total gas and methane yields from the treatment of a distillery wastewater occurred at loading rates of 4.75 and 0.33 kgVS m^{-3} d^{-1} respectively. Overall, a parabolic relationship was observed between gas production rate and organic loading rate with a peak for methane production at 7.66 kgVS m^{-3} d^{-1}. Comparison of mesophilic (37 °C) and thermophilic (53 °C) operation of an MBR treating a wool scouring waste water showed peak gas production rates of c0.20 and c0.25 Nm^3CH_4 kg TOD^{-1} at loading rates of 10 and 20 kg TOD $m^{-3}.d^{-1}$ respectively. The methane content remained at around 80% throughout.

4.4.3 Biomass

Kataoka *et al.* (1992) examined the characteristics of the biomass in anaerobic MBRs by colony forming curve analysis, revealing that bacterial populations for treating sewage are slower growing than for some industrial wastes. This has been explained by the large amounts of cellulosic materials that are generally present in sewage, hydrolysis of which is known to be the rate liming step (Klass, 1984). Analysis of the microbial population in an MBR treating brewery waste revealed a shift in the dominant methanogenic group from *Methanococcus* at start up to medium rods at the end of the study. Although the proportion of methanogens only increased slightly during the study, the number of viable organisms increased sharply resulting in a 6 fold increase in their metabolic activity (Ince *et al.*, 1998). However, Harada *et al.*, (1994) reported on a deterioration in methanogenic activity during operation, attributing this to a change in the balance of net growth with autolysis due to the long sludge ages and low sludge loading rates.

4.4.4 Flux

All the anaerobic MBRs are sidestream in configuration. This is necessary as the lack of air bubbling in the reactor means that fouling amelioration must be accomplished by liquid recirculation via a pump. Excellent mixing conditions between raw incoming feed and biomass have been reported for a MBR treating maize processing effluent at a recycle ratio of 1:6. This represents complete

reactor content recycle every 19 h and a power input of 14 W m^{-3}, around twice the recommended value from the US EPA (Ross *et al.*, 1992). However, examination of the effects of pumping on the activity of methanogenic sludges has shown that 50% of the activity is lost at a pumping rate of 20 cycles (i.e. 20 reactor volumes per day) dropping to 10% at 100 cycles (Brockmann and Seyfried, 1997). Similarly, Choo and Lee (1996a, b) reported on a bench scale MBR where the biomass concentration dropped from 3 to c0.3 gVSS l^{-1} in the first 20 d of operation as compared to a stable 12 gVSS l^{-1} in a conventional reactor. The loss of activity was attributed to cell lysis caused by mechanical shear from the pump.

In the treatment of a brewery waste, Strohwald and Ross (1992) investigated the effects of crossflow velocity and pressure on membrane performance. At a mixed liquor concentration of 30,000 mg l^{-1} no increase in flux was observed when increasing the TMP from 140 to 340 kPa. Similar pressure independency has been demonstrated in other systems at 200kPa (Ross *et al.*, 1990) and represents the transition between low and high pressure operation (Beaubien *et al.*, 1996). In this region crossflow velocity and solids concentration are known to control performance. In the case of a brewery waste, increasing crossflow velocity from 1.5 m s^{-1} to 2.6 m s^{-1} increased the stable flux from c20 l m^{-2} h^{-1} to 45 l m^{-2} h^{-1} (Strohwald and Ross 1992). Furthermore, at a crossflow velocity of 1.5 m s^{-1} a rapid decrease in flux was observed once the biomass concentration had exceeded 20,000 mg l^{-1}, from 33 l m^{-2} h^{-1} at 22,000 mg l^{-1} to 14 l m^{-2} h^{-1} at 50,000 mg l^{-1}. However, in another study, a critical solids concentration of 40,000 mg l^{-1} was observed at a crossflow velocity of 2 m s^{-1}, below which no effect was noticed (Ross *et al.*, 1990). This correlated well with the crossover point from turbulent to laminar flow at the given velocity. Similarly, in constant flux systems with intermittent suction operating on biomass concentrations below 18,000 mg l^{-1}, Wen *et al.* (1999) reported pressure increase rates of 28 kPa d^{-1} for a constant flux of 5 l m^{-2} h^{-1} and 8.3 kPa d^{-1} for a constant flux of 10 l m^{-2} h^{-1}. However, at higher mixed liquor concentrations pressure increase rates are significantly higher. Cadi *et al.*, (1994) recorded a rate of pressure increase of 33 kPa d^{-1} while operating at biomass concentrations between 40,000 and 75,000 mg l^{-1}. Relationships between stable flux and sludge concentration have been reported as linear at <20 mg l^{-1}, (Beaubien *et al.*, 1996) and semi-logarithmic at high mixed liquor concentrations, (Pillay *et al.*, 1994). Rheological analysis of the sludge with increasing concentration revealed it to be pseudoplastic thixotropic (Ross *et al.*, 1990) which is in agreement with findings for aerobic systems at similar concentrations (Rosenberger *et al.*, 1999).

In a series of bench scale fouling tests with a distillery waste the final flux rate was independent of the material or pore size of the membrane used;

although a slight difference in the flux decline profiles was observed (Choo and Lee, 1996a, b). Detailed analysis of the fouling revealed that although fine colloids represented only 5% of the total solids, they accounted for 80% of the total resistance to filtration at around 255×10^{11} m^{-1}. This suggested that filtration performance could be significantly improved by either degrading or agglomerating the fines. This has been demonstrated by powdered activated carbon addition into the bioreactor (Choo et al., 1999). The effect of membrane material and pore size was also investigated with respect to internal fouling. A minimum in fouling index was observed at pore sizes of 0.1 μm. This was attributed to the fact that at higher sizes, macro organics can foul the internal pores and at lower sizes the membrane has too high a natural resistance. A material dependency was seen at higher pore sizes, with a fluoropolymer membrane demonstrating the greatest degree of fouling, although it was the most hydrophobic, indicating that hydrophobicity alone may not be sufficient to describe membrane fouling.

Investigations into the effects of biosolid fouling have shown three phases of flux decline: firstly, a rapid exponential decline over the first 3 d, then a gradual further reduction and finally a steady-state flux. In one study on the fouling from an anaerobic digester treating a distillery waste, 87% of the total resistance was accounted for by surface fouling. The initial flux decline was strongly linked to a decreasing particle size distribution, with measurement of specific cake resistance increasing from 3.9 to 6.4×10^{15} m kg^{-1} as the mean particle size decreased from 20 to 3 μm. Elmaleh and Abdelmoumni (1998) showed that operation without fouling could be achieved at high crossflow velocities because the fouling was derived from biosolids. A flux of 120 l m^{-2} h^{-1} was generated at a crossflow velocity of 3.5 m s^{-1} and a trans-membrane pressure of 0.5 bar. A further flux improvement up to 180 l m^{-2} h^{-1} was achieved by the inclusion of baffles.

Kimura (1991) reported on the cleaning cycles of a range of different membrane configurations used for anaerobic treatment for a variety of wastes. In most cases the membranes were cleaned at short intervals by flushing or backwashing, although in some plate and frame systems operation occurred for long periods of time without cleaning. Flow stopping (Choo and Lee, 1996a) and pH changes (Cadi et al., 1994) have also been shown to greatly affect the nature of the gel layer and hence flux. However, in one study, fouling was attributed to the formation of struvite as well as biosolids (Choo and Lee, 1996b). Periodic acidic backfeeding doubled the flux for a polymeric membrane but only caused initial improvement in ceramic membrane (Choo et al., 1999). This was due to the relatively strong affinity between the ceramic membrane

and the precipitate. A slight additional benefit was reported by selectively removing the ammonium ions in a combined dialysis/zeolite unit.

The work discussed is consistent with the overview of Beaubien *et al* (1996) who demonstrated two clear modes of operation. The first is a low pressure mode where only pressure and solids concentration affect flux. Hysteresis effects have been shown in this mode, whereby the flux pressure response is different with advancing and receding pressures. In the high pressure mode, operation becomes pressure independent but shear stress is now a controlling factor. Further, Beaubien *et al.*, (1996) defined an optimum pressure that balances the benefits of low pressure, low fouling operation with the need to maximise the flow. The optimum pressure was shown to be independent of solids concentration but increased in a linear manner with crossflow velocity. At 1.5 m s^{-1} the optimum pressure was ca. 45 kPa whereas at 3.4 m s^{-1} the pressure increased to 100 kPa.

Table 4.7: Biological performance of anaerobic MBRs treating wastewater

Wastewater		V (m³)	HRT (h)	θx (d)	Load (kg COD m⁻³ d⁻¹)	BOD^a, COD^b, TODL (mg l⁻¹) In	Out	MLSS (kg m⁻³)	Yield (kg VSS kg COD⁻¹)	Gas (m³ CH₄ kg COD⁻¹)	Reference
Palm oil mill	SS	0.05	67	161	14.2	39910[b]	2710[b]	50.7	—	0.28	Fakhru'l-Razi and Noor, 1999
Palm oil mill	SS	0.05	75.6	77	21.7	68310[b]	5390[b]	56.6	—	0.24	
Distillery	SS	2.4	79.2	—	11	37000[b]	2600[b]	50	—	—	Ross et al., 1990
Dairy	SS	0.19	170	25	8	58465[b]	722[b]	24[v]	0.12	0.28	Li et al., 1985
Dairy	SS	0.19	105	30	8.2	35175[b]	270[b]	22.4[v]	0.09	0.29	
Brewery	T/SS	0.05	12	—	15	6700[b]	268[b]	50	—	0.16	Strohwald and Ross, 1992
Synthetic	PF/SS	0.01	120	—	1	5000[b]	<80[b]	15[v]	—	—	Harada et al., 1994
Brewery	T/SS	0.12	60–100	—	<28	85000[b]	2550[b]	50[v]	0.038	0.28	Ince et al., 1998
Maize	T/SS	2.6	124	—	2.9	15000[b]	400[b]	21	—	—	Ross et al., 1992
Wool scour	HF/SS	4.5	—	—	<50[l]	102400[l]	11264[l]	—	—	0.2[l]	Hogetsu et al., 1992
Kraft pulp	SS	0.02	—	—	10.7[l]	10700[l]	1070[l]	7.6	—	1.08[l]	Minami et al., 1991
Synthetic	T/SS/C	0.075	135	52	2[b]	9700[b]	300[b]	8.1	—	0.37	Caid et al., 1994

[101]

Table 4.8: Membrane performance of anaerobic MBRs treating wastewater

Wastewater	Configuration	V (m³)	HRT (h)	Pore size (μm or Da)	Cross flow (m s⁻²)	Pressure (bar)	Final or mean flux (l m⁻² h⁻¹)	Specific flux (l m⁻² h⁻¹ bar⁻¹)	Membrane area (m²)	Reference
Palm oil mill	SS	0.05	72	200,000	2.3	1.5	26 31	17–21	—	Fakhru'l—Razi and Noor, 1999
Brewery	T/SS	0.05	—	40,000	1.5	1.8	18	10	0.44	Strohwald and Ross, 1992
Pulp and paper	PF/SS	7	17	200,000	1	0.45	12.5	27.7	20	Kimura, 1991
Sewage	CA/SS	5.4	6.5	15,000	0.7	1.5	15.8	10.5	100	
Wheat starch	HF/SS	—	—	0.1	0.9	0.79	16.2	20.5	54	
Distillery	PF/SS	8	15	200,000	1	1.3	13.7	10.5	12	
Night soil	CA/SS	0.4	19	15,000	0.9	1.2	6.6	5.5	30	
Synthetic	T/SS	0.01	—	0.14	3.5	0.5	120	240	—	Elmaleh and Abdelmoumni, 1998
Municipal	T/SS	1.8	336	UF	2	2	50	25	—	Pillay et al, 1994
Distillery	PF/SS	0.004	360	20,000	0.57	2	5	2.5	0.36	Choo and Lee, 1996a, b
Egg processing	—	80	32	—	1.8	2.6	23	8.8	—	Brockmann and Seyfried, 1997
Synthetic	T/SS/C	0.007	6	0.2	2	0.4	17	40	0.2	Cadi et al, 1994
Wool scouring	HF/SS	4.5	—	13,000	—	2	14	7	0.2	Hogetsu et al, 1992

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5

Membrane aeration bioreactors and extractive membrane bioreactors

5.1 INTRODUCTION

Research into membrane aeration bioreactors (MABR) and extractive membrane bioreactors (EMBR) has been driven by the need to enhance the performance capabilities and the scope of application of biological wastewater treatment processes. Both processes exploit a membrane's ability to separate two distinct phases, while allowing transport of components from one phase to the other, in order to optimise conditions within the bioreactor for enhanced microbial degradation of wastewater pollutants.

Due to the low solubility of oxygen in water the treatment of high oxygen demanding wastewaters by aerobic processes is often limited by insufficient oxygen when using atmospheric air. By switching from air to high purity oxygen the rate of oxygen mass transfer can be significantly increased. However, conventional oxygenation devices have high power requirements associated with the need for high rate mixing and cannot be used in conjunction

with biofilm processes. Biofilm processes are advantageous as they enable the retention of high concentrations of active bacteria. The MABR process uses gas permeable membranes to directly supply high purity oxygen without bubble formation to a biofilm. The MABR is an attractive alternative to conventional technologies for the treatment of high oxygen demanding wastewaters.

A major problem encountered when attempting to biologically treat industrial wastewaters is the presence of high concentrations of inorganic material, such as acids, bases and salts residual from industrial processes. These may deter the growth and activity of a microbial population otherwise capable of degrading any toxic compounds present. By using tubular silicone membranes to selectively extract chlorinated aromatic compounds from polar and ionic compounds, the extractive membrane bioreactor provides suitable conditions for biodegradation of priority pollutants originally discharged in hostile industrial wastewaters.

This chapter critically reviews the development of these two generic membrane bioreactor technologies.

5.2 MEMBRANE AERATION BIOREACTORS

Wastewater treatment processes using high purity oxygen have a greater volumetric degradation capacity compared to conventional air aeration processes. As a result of the high cost of oxygen, processes need to achieve high oxygen utilisation efficiencies and high standard aeration efficiencies (Chapter 3). The two commercial pure oxygen activated sludge processes, surface oxygenation of a multi-stage sealed reactor and venturi oxygenation, have high oxygen transfer efficiencies (50–90%). Furthermore, the contact loop in the venturi process is operated at high pressures to ensure high saturation concentrations. However, values for the utilisation of oxygen have not been reported, and though transfer efficiencies may be high oxygen molecules are subject to both mass transfer and diffusional limitations before utilisation by the microbial population. Also, due to the high degree of mixing in the surface oxygenation process and the high rate recirculation of the venturi process, standard aeration efficiencies of 2.8 to 5.5 kgO_2 kWh^{-1} are achieved, only 1–3 times greater than air aeration processes. Alternatively, the MABR process uses gas permeable membranes to supply high purity oxygen directly to a biofilm without bubble formation. In the MABR process the capability of biofilm to retain high concentrations of active bacteria is coupled with the high rate transfer of oxygen to the biofilm. Therefore the MABR is an attractive alternative to conventional processes for the treatment of high oxygen demanding wastewaters.

5.2.1 Membrane aeration bioreactor fundamentals

5.2.1.1 Membrane Aeration in the MABR Process

Dense gas permeable, hydrophobic porous and composite membranes are used to transfer oxygen to degradative bacteria present in the bioreactor without bubble formation (Figure 5.1). The membrane usually also acts as support media for biofilm development at the membrane/liquid interface. Wastewater flows over the outer surface of the biofilm and counter-diffusion of oxygen and pollutant occurs. Oxygen is utilised during the biodegradation of pollutants within the biofilm. However, though high purity oxygen is usually used in MABR processes, some investigations have been carried out using atmospheric air (Brindle *et al.*, 1996a).
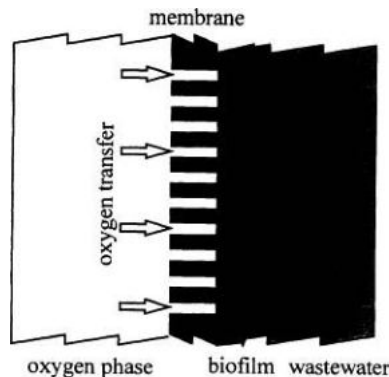
Figure 5.1. Schematic of the MABR process with a biofilm attached to the wastewater side of a hydrophobic porous membrane. Oxygen is transported through the pores of the hydrophobic membrane material into the biofilm. Organics and nutrients are transferred from the wastewater to the biofilm. Transfer of dissolved and gaseous metabolites from the biofilm to the wastewater also occurs.

Plate and frame, tubular and hollow fibre membrane configurations have been used in MABR processes (Table 5.1). The specific surface area of such membranes ranges from a 19 $m^2\,m^{-3}$ plate and frame unit to a hollow fibre unit of 5108 $m^2\,m^{-3}$; far greater than in conventional attached-growth bioreactors (Pankhania *et al.*, 1994; 1999; Timberlake *et al.*, 1988). Research has focused

on hollow fibres, with the gas phase on the lumen side and the wastewater on the shell side of the fibres. These provide a high surface area for oxygen transfer and biofilm formation while occupying a relatively small volume within the bioreactor (2–4%). Pressurised hollow fibre and tubular membranes have been investigated in dead-end and flow-through modes of operation. The evacuation of carbon dioxide from the bioreactor is a benefit of flow-through operation, though no quantitative work to determine removal rates has been undertaken (Cote *et al.*, 1988; 1989; Kniebusch *et al.*, 1990).

Though 100% oxygen transfer efficiency can be achieved when using hydrophobic dead-end hollow fibres their use has been avoided until recently. This was due to condensate formation in the lumen reducing the active membrane surface area for oxygen mass transfer (Cote *et al.*, 1988; 1989). Chemical treatment of the dead-ends of these hollow fibres provides means for condensate to escape (Pankhania *et al.*, 1994; 1999). Since the lumen gas partial pressure is independent of the depth at which the membrane is submerged, large transfer driving forces in shallow depths can be achieved when using dead-end hollow fibres (Cote *et al.*, 1988; 1989). The non-biological fouling and loss of performance of dead-end porous hollow fibres due to iron oxidation, absorption of free oils and greases into pores, surfactants, suspended solids and fibre tangling has been reported (Semmens *et al.*, 1993). Fluidised hollow fibre bundles, with individual fibres independent of one another, are known to be less susceptible to clogging (Ahmed *et al.*, 1992a; b).

Table 5.1: Membranes used in MABR applications

Membrane Configuration	Specific surface area (m² m⁻³)	Outside diameter (mm)	Wall thickness (μm)	Pore size (μm)	Reference
Dead-end Teflon hollow fibre	550	0.8	0.23		Yeh et al., 1978
Flow-through polytetrafluoroethylene hollow fibre	60	3.5	0.5	2.0	Suzuki et al., 1993
Plate & frame polytetrafluoroethylene between 2 nylon layers (Gore-tex)	19		0.025	0.2	Timberlake et al., 1988
dead-end polypropylene hollow fibre	5108	0.028			Pankhania et al., 1994
Flow- through polyetherimide			0.026	0.05	Kniebusch et al., 1990
Plate & frame				—- 10.0	
Plate & frame silicone	34	4.0	0.5	—	Debus et al., 1992
Flow-through hollow fibre silicone & fibrous support layer	62	1.0	0.5	—	Hirasa et al., 1991

Oxygen diffusion through dense gas permeable membrane material can be achieved at high gas pressures without bubble formation. Work using dense polymers has largely focused on tubular silicone membranes; these have high oxygen permeabilities, and are very resistant to chemical and mechanical abrasion (Debus et al., 1992). It has previously been suggested that biofilm formation does not occur on silicone membranes when used to supply oxygen, due to either the hydrophobic nature of the silicone or biomass poisoning by high oxygen concentrations at the membrane wall (Cote et al., 1988; 1989; Wilderer et al., 1985). However, several investigations have demonstrated that silicone is a suitable material for biofilm attachment during diffusion of oxygen through the membrane material (Debus et al., 1992). Compared to microporous membranes, silicone membranes have thicker walls and a greater resistance to oxygen transport (Ahmed et al., 1992a; b).

In hydrophobic microporous membranes the pores remain gas filled; oxygen is transported to the shell-side of the membrane through the pores by gaseous diffusion or Knudsen flow transport mechanisms. The partial pressure of oxygen is kept below the bubble-point to ensure bubble-less oxygenation when using clean membrane materials (Ahmed et al., 1992a; b; Semmens 1991). Semmens et al. (1993) described a composite porous hollow fibre (pore diameter 0.04–1.0 μm) with a very thin (< 1 μm) dense polymer layer that can provide bubble-less aeration at gas partial pressures of up to 6.95 bar.

In laboratory-scale experiments surveyed by Cote et al. (1988; 1989), the mass transfer coefficient ranged from between 0.1 g O_2 m^{-2} h^{-2} bar^{-1} for a dense polyethylene with no hydrodynamic mixing to 11.5 g O_2 m^{-2} h^{-2} bar^{-1} for porous polysulphone in an intensely mixed clean water. In clean systems, i.e. without a membrane attached biofilm, the liquid boundary layer has a significant influence upon the overall oxygen mass transfer, with the shear velocity at the membrane/liquid interface a key operational parameter (Cote et al., 1988; 1989; Kniebusch et al., 1990). However, the wall thickness of dense membranes can affect the transport of oxygen (Wilderer, 1995).

In the MABR process the oxygen transfer rate (OTR) can be enhanced by increasing either the mass transfer driving force (C^*-C), the membrane surface area (a) or the overall mass transfer coefficient (K). For a given set of operating conditions, increasing the membrane surface area yields greater oxygen transfer rates without requiring more power and therefore also improves the standard aeration efficiency (SAE). Maintaining a high mass transfer driving force can be achieved by supplying oxygen at higher partial pressures (P_T) or keeping the dissolved oxygen concentration at zero (C) (Chapter 3). In conventional aeration devices the oxygen feed pressure (P_T) is limited by the atmospheric and hydrostatic conditions (WPCF 1988). With dead-end hollow fibres intra-membrane oxygen pressures as high as approximately 6 bar can significantly increase the driving force for oxygen mass transfer (Ahmed et al., 1992a; b).

In the MABR process the microbial community in the biofilm is the desired destination of the oxygen supplied. Control of the intra-membrane oxygen to match the oxygen requirement (Q_o) ideally results in all the oxygen being consumed within the biofilm and the maximum possible concentration difference (C^*–C) existing across the biofilm (Section 3.5, Chapter 3). Thus, the low solubility of oxygen in water and the stagnant boundary layer resistance to mass transfer becomes less significant during MABR operations. The high oxygen transfer rate is due to oxygen utilisation within the biofilm maintaining the high concentration gradient.

A 100% oxygen transfer efficiency can be achieved using dead-end hollow fibre membranes in the MABR process (Ahmed et al., 1992a; 1992b). 100% oxygen utilisation efficiency (OUE) is possible because all the oxygen in the

fibre lumen transfers to the biofilm. A pure oxygen process that efficiently utilises oxygen has a lower operating cost than a process where oxygen is wasted. A laboratory MABR unit operated for 172 d nitrified an ammonia rich synthetic wastewater at different ammonia loading rates and oxygen supply regimes. For 41 d the oxygen supply matched the oxygen utilised by the bacterial population within the biofilm and 100 ± 10% OUE was achieved. The high OUE was due to the proximity of the biofilm to the oxygen source, the activity of bacterial population within the biofilm, a 100% OTE, and by interfacial oxygen mass transfer (Brindle *et al.*, 1998; Pankhania *et al.*, 1999).

5.2.1.2 MABR Biofilms

Microbial communities in wastewater treatment processes exist as aggregated flocs suspended in the bulk liquid and/or within films immobilised on solid surfaces within the reactor. Biofilms enable the retention of high concentrations of active bacteria. They are complex matrixes that enclose microbial communities, polymeric substances excreted by the bacteria, entrapped non-biological solids, substrates, metabolites and interior pores and channels. Microorganisms within biofilms have a longer residence time than cells within suspended flocs; the later being continuously removed from the reactor with the bulk liquid. Biofilms enable the development of microbial species that have growth rates slower than the hydraulic retention time of the process. Therefore, nutrient removal and the degradation of inhibitory and complex pollutant compounds, which require slow growing microbial communities, can be achieved in biofilm processes without the long HRTs or high biomass recycle ratios required in suspended floc processes (Bishop *et al.*, 1995; Loosdrecht *et al.*, 1995).

Biofilms are diverse and complex systems, with the lack of generality leading to the belief that biofilm behaviour is a function of reactor type (Cao *et al.*, 1995). The environmental factors and biofilm characteristics that influence biofilm behaviour are interrelated, complex and not completely understood. Environmental factors particularly pertinent to MABR biofilms include the nature and concentration of growth substrate(s), liquid phase velocity at liquid/biofilm interface, boundary layer thickness, nature of support media and transfer and entrapment of non-biological solids into the biofilm. Important biofilm characteristics include microbial kinetics and speciation, interspecies co-operation and competition for available substrate(s), production of extracellular polymer substances, biofilm morphology thickness and density and detachment forces. These factors will not be discussed in-depth, although their influence upon the operation and performance of MABRs has been referred to where necessary.

In MABRs the membrane used for oxygen mass transfer usually also provides support for biofilm attachment at the membrane/liquid interface (Figure 5.1). However, as previously stated, some MABR processes have been investigated with additional/alternative non-permeable support material for biofilm growth. To prevent biofilm growth and bubble development at the membrane/liquid interface and to reduce the effect of the liquid boundary layer on oxygen transfer into the bulk liquid, high shear velocities would be required. Therefore, operating an MABR with a suspended biomass or a biofilm attached to a non-permeable support would result in an increase in the power used and a reduction in the SAE.

Membrane attached biofilms are in intimate contact with the oxygen source, with direct interfacial transfer and utilisation of oxygen within the biofilm. Thus, in such biofilms oxygen from the membrane and pollutant substrate(s) from the bulk liquid transfer across the biofilm in counter-current directions (Figure 5.2). In conventional biofilm processes, including biofilms attached to non-permeable material within MABRs, both oxygen and pollutant substrate(s) travel co-currently through the biofilm, from the liquid/biofilm interface to the biofilm/support interface (Figure 5.3).
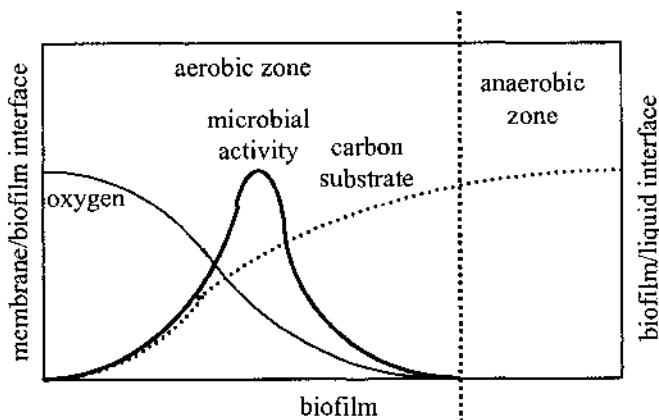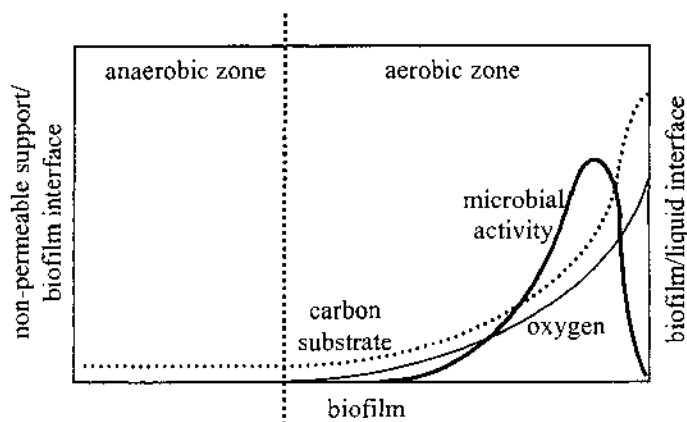

Figure 5.2. Simplified representation of the concentration profiles of oxygen, carbon substrate and microbial activity in a membrane attached biofilm. (This diagram does not represent the profiles of all membrane attached biofilms.)

This difference between a membrane attached biofilm and a biofilm grown on a non-permeable support material results in a significant difference in the oxygen and pollutant substrate profiles through the biofilm, microbial activity,

location of the active layer and microbial speciation of the two biofilms (Casey et al., 1999; Debus 1995; Essilia et al., 1997; Timberlake et al., 1988; Wilderer 1995). In a membrane attached biofilm the highest concentration of oxygen is at the membrane/biofilm interface, while the highest carbon substrate concentration is at the biofilm/liquid interface. This can result in the location of highest microbial activity near the centre of the biofilm, where both oxygen and carbon are plentiful. In this biofilm the anaerobic conditions exists near the biofilm/liquid interface (Figure 5.2). If the biofilm is thin high oxygen intra-membrane pressures exist complete oxygen penetration of the biofilm occurs and aerobic conditions prevail. In a conventional biofilm the highest concentration of oxygen and carbon substrate is at the biofilm/liquid interface (Figure 5.3). This results in the location of highest microbial activity near biofilm liquid interface. In this biofilm anaerobic conditions exist near the non-permeable support/biofilm interface. Furthermore, as oxygen can be supplied at high intra-membrane pressures, complete oxygen penetration of biofilms greater than 1mm thick have been demonstrated. Micro-electrode examination of a membrane attached biofilm revealed that switching from air to pure oxygen increases the oxygen penetration, biofilm oxygen concentration and the biofilm respiration rate (Wilderer 1995).

Figure 5.3. Simplified representation of the concentration profiles of oxygen, carbon substrate and microbial activity in a biofilm attached to a non-permeable support material. (This diagram does not represent the profiles of all biofilms attached to non-permeable support material.)

Biofilm thickness and morphology vary with each MABR investigation (Table 5.2). During the nitrification of a synthetic solids-free wastewater a relatively smooth thin biofilm existed along the lengths of hollow fibre hydrophobic membranes (Brindle et al., 1998; Brindle et al., 1996b). A scanning electron micrograph showed a biofilm of approximately 400-500μm thick attached to a similar hollow fibre membrane during the treatment of a high strength synthetic wastewater (Pankhania et al., 1994). Scanning electron micrographs of membrane attached biofilm have also revealed that bacteria will inhabit the membrane pores with a diameter of 10 μm; in this case the location of the oxygen and wastewater interface very close to the bacteria (Rothemund et al., 1994). Internal pores and channels within membrane attached biofilm may exist that would result in the mass transfer of components within the matrix.

Table 5.2: Characteristics of membrane attached biofilms

Effluent type	Biofilm support	Biofilm thickness (mm)	Biological culture	Comment
Primary sewage[1]	Gore-tex	≈ 1	mixed, aerobic & anaerobic	Considerable spatial variation in biofilm thickness.
Primary sewage[2]	polypropylene	≈ 1-4	mixed, aerobic & anaerobic	Density of biofilm 80 to 105 kg m^{-3}.
Synthetic wastewater[3]	silicone tubing sheathed with fibrous support	≈ 1(on support surface)	seeded with nitrifier enriched activated sludge	Spatial distribution of heterotrophs, nitrifiers and denitrifiers within biofilm
Xylene synthetic wastewater[4]	silicone tubing	≈ 2	seeded with acclimated bacterial population from activated sludge	Detachment of biofilm led to development of suspended bacterial population.
2,4 dichlorophenoxyacetate[5]	polyether imide	≈ 0.2	seeded with pure culture of *Alcaligens eutrophus*	a pore size of 10 μm at the membrane/biofilm interface enabled bacterial colonisation.

[1] Timberlake et al., 1988; [2] Osa et al., 1997; [3] Yamagiwa et al., 1998; [4] Debus, 1995; [5] Kniebusch et al., 1990

5.2.1.3 MABR Reactor Configurations & Operation

Membrane aeration bioreactors are operated with a biofilm attached to the surface of the membrane at the membrane/liquid interface (Figure 5.1), though some processes also incorporate a non-permeable biofilm support media to increase the volumetric biomass concentration within the reactor (Tables 5.3 and 5.4). Apart from the different membrane configurations (i.e. hollow fibre, tubular, plate and frame) reactors vary with regard to the method and degree of liquid phase mixing, incorporation of additional non-permeable biofilm support media, membrane unit integral or external to the bioreactor and continuous or batch operation.

In conventional aerobic biological wastewater treatment processes liquid mixing is essential to ensure that pollutant removal rates are not mass transfer limited. These processes usually rely upon air bubble aeration to provide the necessary mixing as well as oxygen for microbial utilisation. In the MABR process oxygen is transferred without forming bubbles and therefore cannot be utilised to mix the bulk liquid. In laboratory scale MABRs liquid phase mixing has been achieved using recirculation pumps, impellers, magnetic stirrers, nitrogen or air sparging (Tables 5.3 and 5.4). Owing to the low solubility of oxygen in water, MABR processes that require the oxygen to be transferred beyond the membrane attached biofilm need intensive liquid mixing to achieve high rate and efficient oxygen utilisation. Therefore, MABR processes that incorporate additional non-permeable support media will either suffer lower rates and efficiencies of oxygen utilisation and/or lower standard aeration efficiencies compared to membrane attached biofilm MABR processes. However, the materials used to increase the surface area for biofilm growth (woven fabric, perforated plastic tubes, and blasted clay) are considerably cheaper than the membranes used and thus the capital cost would be less than opting to increase the membrane surface area.

With regard to membrane placement within MABR processes two configuration types have been investigated; one with the membrane and bioreactor integrated into the same unit and the other with the membrane external to the bioreactor and incorporated within the recirculation line (Table 5.4). In most investigations the membrane is integral to the bioreactor. However, the external membrane aeration of a sequencing batch biofilm reactor was carried out using tubular silicone membranes with an attached biofilm to degrade volatile organic carbons and thus prevent them transferring to the gas phase within the tubes (Debus 1995). In this process there is a need for high rate recirculation between the biofilm reactor and the membrane aeration unit; therefore reducing the SAE of the process.

Table 5.3: Reactor configuration and operation of MABR with only membrane attached biofilm

Membrane	Reactor configuration	Method of mixing	Degree of mixing	Mode of operation
Dead-end hydrophobic porous membrane[1]	Tubular column	Recirculation pump	Completely mixed	Continuous
Dead-end hydrophobic porous membrane[2]	Tubular column	None	Laminar	Continuous
Plate and frame PTFE[3]	Cylindrical vessel	Impeller	Completely mixed	Continuous
Tubular polypropylene[4]	Cylindrical vessel	Magnetic stirrer	Completely mixed	Continuous
Plate and frame Gore-tex[TM5]	Cylindrical vessel	Nitrogen sparged	Completely mixed	Continuous
Plate and frame polyetherimide[6]	Cylindrical vessel	Recirculation pump	Completely mixed	Continuous
Reinforced coiled silicone tubing[7]	Cylindrical vessel	Recirculation pump	Completely mixed	Batch
Coiled silicone tubing[8]	Cylindrical vessel	Magnetic stirrer	Completely mixed	Continuous

1 Brindle *et al.*, 1998; Brindle *et al.*, 1996; Brindle *et al.*, 1999; Pankhania *et al.*, 1994; Pankhania *et al.*, 1999; 2 Brindle *et al.*, 1999; 3 Osa *et al.*, 1997; 4 Osa *et al.*, 1997; 5 Timberlake *et al.*, 1988; 6 Kniebusch *et al.*, 1990; 7 Wobus *et al.*, 1995; 8 Debus 1995; Debus *et al.*, 1992.

Table 5.4: Reactor configuration and operation of MABR with membrane attached biofilm and biofilm attached to additional non-permeable support material

Membrane	Additional support media	Int.[a]	Reactor geometry	Method & degree of mixing	Mode of operation
Reinforced tubular silicone[1]	perforated plastic tubes (25mm diameter)	✓	Horizontally baffled rect. Tank	None/ laminar	Continuous
Coiled tubular silicone[2]	granular activated carbon	✓	Cylindrical vessel	Recirculation pump/ completely mixed	batch
Coiled tubular silicone[3]	granular activated carbon	✗ ✓	Cylindrical vessel	Recirculation pump/ mixed	batch
Tubular silicone[4]	Blasted clay	✗	Cylindrical vessel	Recirculation pump/mixed	batch
Tubular silicone[5]	Woven fibre sheath over silicone tubes	✓	Cylindrical vessel	Air sparged	Continuous

[a] ✓, Membrane and additional support media integrated into one reactor. ✗, Membrane external to the reactor housing the additional support media. [1]Richter et al., 1994; [2]Chozick et al., 1991; [3]Kolb et al., 1995; [4]Dollerer et al., 1996; [5]Yamagiwa et al., 1994;

Yamagiwa et al., (1994) tried to overcome the surface area limitations of MABRs operated with only membrane attached biofilm and the oxygen mass transfer limitations of MABRs operated with additional non-permeable biofilm support media, by sheathing individual tubular silicone membranes with a close fitting woven fibrous material. The biofilm initially developed at the membrane/woven sheath interface and continued to grow through and beyond the woven sheath. Thus, the process was able to achieve simultaneous organic carbon removal and nitrification.

Membrane aeration bioreactors have been operated in continuous and batch modes. Usually MABRs relying solely on membrane attached biofilms are operated continuously, while processes using an additional support media are batch operated (Tables 5.3 and 5.4). Kolb et al. (1995) described a batch process coupled with a MABR plus an activated carbon bed for the treatment of VOCs in a wastewater. At organic and 2 chlorophenol removal rates as high as 15.5 kg m^{-3} d^{-1} and 20 kg m^{-3} d^{-1} respectively, the activated carbon ensured the rapid reduction of the VOC concentration to below the toxicity limits of the microorganisms at the beginning of each cycle. This was proceeded by microbial

biodegradation of the activated carbon attached VOCs. Colonisation of the activated carbon by the microbial population was shown to have no influence on the absorptive capacity of the activated carbon.

In continuously operated MABR processes control of the biofilm thickness is a key operational consideration. Excessive biofilm accumulation can result in oxygen, pollutant substrate and nutrient mass transfer limitations, excessive membrane fouling, a decline in biomass activity, metabolite accumulation deep within the biofilm, and the channelling of flow in the bioreactor such that steady-state conditions cannot be maintained (Debus *et al.*, 1992; Pankhania *et al.*, 1994; Wanner *et al.*, 1994; Yeh *et al.*, 1978). To operate effectively regular membrane washing, air scouring backwashes, and high shear velocities have been employed to control biomass accumulation (Debus *et al.*, 1992; Pankhania *et al.*, 1994; Rothemund *et al.*, 1994; Yeh *et al.*, 1978). Furthermore, hollow fibre bundles tend to clump together when biofilm growth is excessive. This leads to fibre tangling and a reduction in the membrane surface area available for biofilm growth (Pankhania *et al.*, 1994).

The frequency and intensity of the membrane cleaning procedure depends upon the loading rate of the wastewater, the bacterial yield, the microbial production of extracellular polymer substances and the retention of non-biological solids in the biofilm matrix. No membrane cleaning was required over 172 d during the treatment of an ammonia rich synthetic solids-free wastewater (Brindle *et al.*, 1998). In comparison, a small pilot-plant MABR treating a high strength brewery effluent, which was easily degradable and contained a high concentration of suspended solids, required an almost daily membrane clean (Brindle *et al.*, 1999).

5.2.1.4 Applications

Membrane aeration bioreactors have been used to treat a variety of wastewater types (Table 5.5). However, most investigations show that the process is particularly suited to the treatment of high oxygen demanding wastewaters, biodegradation of volatile organic compounds (VOCs), combined nitrification, denitrification and/or organic carbon oxidation in a single biofilm.

As the MABR process usually uses high purity oxygen supplied at high partial pressures without bubble formation and directly to a membrane attached biofilm, much higher oxygen utilisation rates and efficiencies can be achieved compared to conventional pure oxygen processes. Organic removal efficiencies between 63% to 91% of influent COD at loading rates between 0.06 kg m^{-3} d^{-1} and 33.8 kg m^{-3} d^{-1} have been reported (Debus 1992; Pankania *et al.*, 1994; Timberlake *et al.*, 1988; Wanner *et al.*, 1994; Yeh *et al.*, 1978). A laboratory scale polypropylene dead-end hollow fibre MABR achieved 86% COD removal at a loading rate of 8.94 kg m^{-3} d^{-1} and a 36 min HRT over 5 months with a biomass yield coefficient of 0.41 kg TS kg COD^{-1} (Pankania *et al.*, 1994).

That the MABR process can achieve high organic removal rates and efficiencies without the need for intensive liquid mixing has recently been demonstrated (Brindle *et al.*, 1999; Pankhania *et al.*, 1999). A brewery wastewater, consisting of easily degradable dissolved organics, was treated by a pilot-plant MABR using dead-end composite hollow fibres (Brindle *et al.*, 1999). An organic removal rate and efficiency of 27 kg COD m^{3} d^{-1} and 81% were respectively achieved without mixing, compared to 28 kg COD m^{3} d^{-1} and 88% during completely mixed conditions. It was speculated that the increase in the resistance to organic mass transfer in the liquid phase was overcome by the high organic concentration gradient encountered along the length of the module during plug-flow operation. Successful MABR operated under plug-flow conditions demonstrated that the process can achieve high standard aeration efficiency without a deterioration in removal capability. Though the MABR was not designed to remove suspended solids, 80% and 28% removal efficiencies were achieved respectively under completely mixed and plug-flow conditions. Regular membrane cleaning was required to control the biofilm thickness and the amount of washwater as a percentage of the wastewater treated averaged approximately 9%. Recovery from fibre cleaning was within 0.5 to 2.0 HRTs and the performance during influent shock loads was relatively stable (Brindle *et al.*, 1999).

Nitrification, the oxidation of ammoniacal-N to nitrate-N, requires more oxygen than the oxidation of organic carbon per unit of substrate oxidised. The treatment of a high strength ammoniacal-N synthetic wastewater has been

demonstrated by Brindle and Stephenson (1996b) and Brindle *et al.* (1998). Using a laboratory-scale MABR with dead-end hollow fibre membranes a specific nitrification rate of 13 $kgNH_4$-N $kgSS^{-1}$ d^{-1} was achieved; significantly greater than conventional nitrification processes. Nitrifying bacteria have slow growth rates and as a result no membrane cleaning was required to control the biofilm thickness throughout these experiments.

In conventional aerobic biological wastewater treatment processes VOCs can enter the atmosphere without being biodegraded as a result of air bubbles stripping out the compounds from the bulk liquid. Since no oxygen bubbles are formed during MABR operation the removal of VOCs is limited. However, VOC removal can occur when flow-through membranes are used and the membrane attached biofilm is not able to completely degrade VOCs present in the wastewater. Volatile organic compounds at the membrane/biofilm interface transfer across the membrane material, into the gas phase and exit to the atmosphere. In a laboratory scale silicone membrane bioreactor xylene was no longer transferred to the gas phase once a sufficient biofilm had developed, indicating removal in the biofilm (Debus, 1992; 1995; Wanner *et al.*, 1994). The gas phase assisted removal of VOCs does not occur with membranes operated in dead-end mode as no exit to the atmosphere exists.

Owing to the different locations of the maximum oxygen and pollutant substrate(s) concentrations within a biofilm (Figure 5.2) and the relative thickness of MABR biofilms, significant stratification within the biofilm can exist. Biofilm stratification enables the removal of more than one pollutant type. A biofilm formed on tubular silicone membranes, each sheathed by a fibrous woven support, achieved simultaneous organic carbon removal and nitrification (Yamagiwa *et al.*, 1994; Yamagiwa *et al.*, 1998). Furthermore, the nitrification rate of 0.0022 kg m^{-2} d^{-1} compared well with conventional biofilm processes dedicated solely to nitrification. Nitrifiers were found to dominate the biofilm within the fibrous support, where oxygen concentrations were sufficient for nitrification, while dentrifiers occupied the zone at the biofilm/liquid interface, where anoxic conditions prevailed. Similarly, Timberlake *et al.* (1988) achieved simultaneous nitrification, denitrification and heterotrophic oxidation in a 1mm biofilm attached to a Gore-tex fabric membrane due to the three layered stratification of the biofilm. The high oxygen concentrations coupled with the low organic carbon concentrations near the membrane/biofilm interface encouraged nitrification, an aerobic heterotrophic layer above this facilitated organic carbon oxidation and an anoxic layer near the biofilm/liquid interface supported denitrification.

Table 5.5: MABR applications

Effluent type	Wastewater Components	Pollutant loading rate (kg m^{-2} d^{-1})	Influent pollutant conc. (mg l^{-1})	% pollutant removal	Pollutant removal rate (kg m^{-2} d^{-1})
Primary sewage[1]	TOC	0.003–	70–92	33–50	
	Org-N	0.011	17–27	55–75	
	NH$_4$-N	0.001–0.002 <0.004	14–30		
Primary sewage[2]	COD	0.1–0.6		75–90	<0.18
	Tot. N	<0.1			0.01–0.05
Synthetic[3]	BOD	0.011	200		
	TOC	0.007	114	95	
	Tot. N	0.003	41	50–90	0.002
Synthetic[4]	TOC	0.042[a]	1000	98	
	Tot. N	0.002[a]	59	98	
Synthetic[5]	2,4 dichlorophen-oxyacetate	≈ 0.0003[b]	≈ 2[c]	85	
Synthetic[6]	TOC	0.0048		83	0.0040
Synthetic[7]	NH$_4$-N	0.006	≈ 45–55	83 nitrification 98 N removal	0.005
Brewery effluent[8]	Tot. COD[d]	≈ 0.068	1782 ± 40	83	
	Sus. COD[d]	≈ 0.013	343 ± 49	84	
	Tot. COD[e]	≈ 0.076	2545 ± 69	81	
	Sus. COD[e]	≈ 0.014	465 ± 14	28	

[a]volumetric loading rate (kg m^{-3} d^{-1}); [b]volumetric loading rate (mM m^{-3} d^{-1}); [c] mM l^{-1}; [d] complete-mixed operation; [e] plug-flow operation.

[1]Timberlake et al., 1988; [2]Osa et al., 1997; [3]Yamagiwa et al., 1994; [4]Hirasa et al., 1991; [5]Kniebusch et al., 1990; [6]Suzuki et al., 1993; [7]Brindle et al., 1998; [8]Brindle et al., 1999

5.3 EXTRACTIVE MEMBRANE BIOREACTORS

5.3.1 Extractive Membrane Bioreactor Fundamentals

A major problem encountered when attempting to biologically treat industrial wastewaters is the presence of high concentrations of inorganic material, such as acids, bases and salts. These may deter the growth and activity of a microbial

population otherwise capable of degrading the toxic compounds present. By using tubular silicone membranes to selectively extract chlorinated aromatic compounds from polar and ionic compounds, the extractive membrane bioreactor (EMBR) provides suitable conditions for the biodegradation of priority pollutants originally discharged in hostile industrial wastewaters.

The EMBR process enables the transfer of degradable organic pollutants from hostile industrial wastewaters, via a dense silicone membrane, to a nutrient medium for subsequent biodegradation (Figure 5.5) (Livingston, 1993a; b).
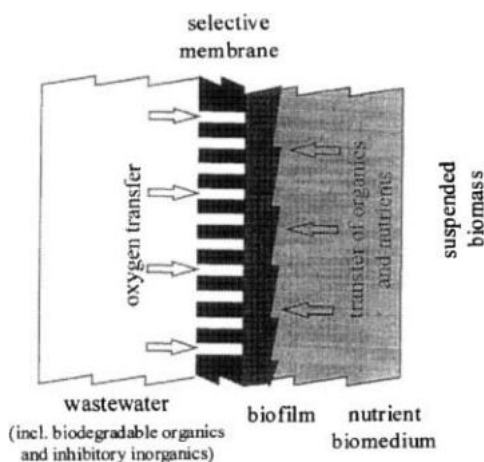
Figure 5.5. Schematic of the EMB process with a biofilm attached to the biomedium side of the dense membrane. Priority organic pollutants selectively diffuse through the membrane material into the biomedium phase. Oxygen and nutrients are transfer from the wastewater to the biofilm and the suspended biomass.

Membranes used for the extraction of pollutants into a bioreactor have been developed from pervaporation by exchanging the vacuum phase with a nutrient biomedium phase where biodegradation mechanisms maintain the concentration gradient needed to transfer organic pollutants present in hostile industrial wastewaters (Lipski et al., 1990; Nguyen et al., 1987). The concentration gradient required to drive the mass transfer of the pollutants from the wastewater through the membrane and into the bioreactor is maintained by the biodegradation of the pollutants in the latter phase. The inorganic composition of the bioreactor is unaffected by the industrial wastewater within the silicone tubes. Hence, the conditions within the bioreactor can be optimised to ensure high biodegradation rates. A biofilm develops on the membrane surface; the

counter-diffusion of organics and oxygen within the biofilm prevents the air stripping of volatile organic compounds (Brookes and Livingston, 1994; Livingston, 1993a; b; 1994).

The extraction and biodegradation of toxic volatile organic pollutants such as choroethanes, chlorobenzenes, chloroanilines and toluene from hostile industrial wastewaters, with high salinity and extremes of pH using EMBRs has been demonstrated. A 2.5 l EMBR successfully removed (>99%) and biodegraded aniline (36 mg l^{-1}), 4-chloroaniline (24 mg l^{-1}), 2,3-dichloroaniline (62 mg l^{-1}), and 3, 4-dichloroaniline (194 mg l^{-1}) from an industrial wastewater with a flow rate of 69 ml h^{-1} and total organic carbon concentration of 168 mg l^{-1} (Brookes and Livingston, 1994; Livingston et al., 1993). Though membrane attached biofilms have been known to reduce mass transfer, Brookes and Livingston (1993a; b; 1994) claim that in this bioreactor the biofilm may actually aid the transfer by effectively maintaining a high localised concentration gradient. Pilot-plants are being trialed at numerous facilities throughout the United Kingdom.

A variation of the EMBR uses an ultrafiltration membrane to separate a biologically sterile nutrient phase from a wastewater phase, with the development of a biofilm at the membrane/wastewater interface (Peys et al., 1997). Pollutant substrates from the wastewater diffuse through the biofilm in a counter-current direction to the nutrients (phosphate buffer and mineral salts) transferred through the membrane. At a feed concentration of 470 mg l^{-1} of 3-chlorobenzoate a removal rate and efficiency of 0.24 kg m^{-2} d^{-1} and 99.5% were respectively achieved. In another variant, Diels et al. (1993) used an EMBR to enhance the biosorption of Cd and Zn for subsequent recovery and concentration. The membrane was of a composite material, polysulphone and ZnO_2, with a heavy metal resistant Alicaligenes eutropheus strain immobilised on the surface.

5.4 MODELLING MABR AND EMBR BIOFILMS

As previously stated, biofilms are diverse and complex systems, leading to practical limitations when it comes to the development of behavioural models. As a result existing biofilm models are equally diverse, with considerable variation in scope. However, most models tend to be one-dimensional and usually predict concentration profiles along the reactor length. Some models also include a second macro-dimension such as reactor hydrodynamics (Vayenas et al., 1994). Numerous biofilm models that consider single growth limiting substrates on non-permeable support media have been developed (Lee et al., 1986; Rittmann, 1982; Suidan, 1986; Suidan et al., 1985). Similar models have been developed for membrane attached biofilms (Beyenal et al., 1994).

Models for mixed culture biofilms that assess the influence of population dynamics on biofilm behaviour have been developed for a single substrate (Rittmann *et al.*, 1992) and multiple-substrates (Furumai *et al.*, 1992; Gujer *et al.*, 1990). The latter include biofilm thickness predictions, spatial distribution and space competition influences, as well as time dependent changes in species concentration. Biofilm models that incorporate dual growth limiting substrates have been developed for single microbial communities for open channels (Li *et al.*, 1994) for EMBRs (Livingston 1994; Livingston 1993a; b; Pavasant *et al.*, 1996) and MABRs (Casey *et al.*, 1999; Essilia *et al.*, 2000). The modelling of MABR and EMBR processes differs from models developed for biofilms grown on non-permeable support, as the oxygen and the pollutant growth substrates diffuse through the membrane attached biofilm in counter-current directions.

Predictions from a steady state biofilm model for the EMBR process agreed with experimental results, where oxygen and phenol were the growth limiting substrates (Livingston 1993a; b; 1994). This model described the membrane transport, diffusion and reaction of phenol. A dynamic model developed by Pavasant *et al.* (1996) investigated cell growth, decay and detachment of a biofilm degrading 1,2-dichloroethane and successfully predicted experimental results.

The first MABR process models were based on a heterogeneous biofilm model to predicted the biodegradation of xylene (Debus *et al.*, 1992; Gujer *et al.*, 1990). The model, which considered two reaction steps and incorporated an oxygen dependent endogenous decay term, compared well with experimental results and revealed that xylene removal rates were affected by biofilm thickness. A calibrated model was used by Wanner *et al.* (1994) to investigate the spatial distribution and dynamics of a heterotrophobic biofilm degrading xylene. Though not comprehensive, analysis revealed that when oxygen was supplied as air the heterotrophic population inhabited a narrow layer near the membrane/biofilm interface and when high purity oxygen was used the heterotrophic active layer moved closer to the biofilm/liquid interface. More recently, Casey *et al.* (1999) and Essilia *et al.* (2000) have developed MABR models to predict oxygen and pollutant substrate mass transport and reaction using dual limiting substrate models.

The predictive model developed by Essilia *et al.* (2000) is similar to the EMBR steady-state model developed by Livingston (1993a; b; 1994); differing in its incorporation of an expression for boundary layer resistance to mass transfer in cylindrical co-ordinates. The model was developed for both membrane attached biofilms and biofilms attached to non-permeable support media, to investigate the affect of the different mechanisms of oxygen supply on their behaviour. The model illustrated that MABR biofilms have the advantage of avoiding resistance to oxygen mass transfer in the liquid boundary layer,

while conventional biofilms have the advantage of coincidence of oxygen and susbstrate profiles, which results in higher maximum microbial activity values. The mass transport advantage of membrane attached biofilms was shown to be particularly significant under conditions of oxygen limitation as well as low liquid velocities. The model predicted that as MABR biofilms can be operated at significantly greater oxygen concentrations than conventional biofilms, substrate removal was greater throughout the entire range of substrate concentrations and liquid velocities values examined for a moderate biofilm thickness (150 μm).

Casey *et al.* (1999) applied a mathematical model developed by Karel and Robertson (1987) for metabolite production of mammalian cell cultures. The rate limiting processes affecting overall MABR performance were identified as oxygen limitation, carbon substrate limitation and dual substrate limitation. For a given set of intra-membrane oxygen pressures and carbon substrate concentrations, an optimum biofilm thickness was shown to exist which maximises the reaction rate by minimising the diffusion resistance of substrate into the active layer. The location of the active layer was found to vary with the ratio of the carbon substrate loading rate to the intra-membrane oxygen pressure.

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6

Commercial membrane bioreactor processes

6.1 KUBOTA

In 1989 the Japanese Government charged many of their large corporations, including Kubota, with investing time and money in new treatment technologies that had a low footprint and produced a high-quality final effluent with re-use capabilities. Out of this initiative was developed the Kubota flat sheet submerged MBR process.

6.1.1 Process Description

In the Kubota process the membrane units are submerged into an activated sludge tank. In common with all MBR processes, the bioreactor can be operated with high MLSS concentrations (Ishida *et al.*, 1993; Churchouse 1997a; b; 1999). Typically, the activated sludge is maintained in the range 15,000–20,000 mg l^{-1} MLSS. The standard Kubota unit comprises two sections. The top section

contains 150 flat panel membranes slotted into a glass fibre reinforced plastic housing allowing a gap of approximately 7 mm between panels (Figures 6.1 and 6.2).
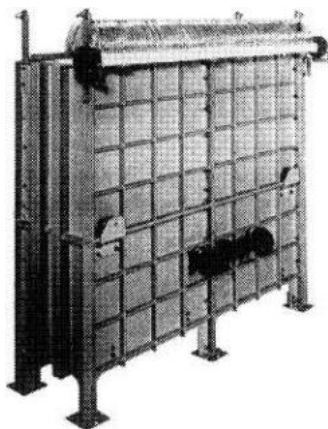
Figure 6.1. Standard glass fibre reinforced plastic Kubota membrane unit containing 150 flat panel membranes.

The lower section of the unit contains a coarse bubble diffuser mounted within a simple matching housing. This supports the top section and channels the bubbles and activated sludge flow between the membrane plates in the upper section. The bubbles released by the lower diffuser section generate an upward sludge crossflow over the membrane surface of approximately 0.5 m s^{-1}. This crossflow minimises fouling and allows low pressure gravity filtration of the treated effluent into the membrane panel and from there to the collecting manifold. Each membrane panel comprises of a solid acrylonitrile butadiene styrene support plate with spacer layer between it and an ultrasonic welded flat sheet membrane on both sides. Each panel has 0.8 m^2 of membrane of a nominal pore size 0.4 μm.
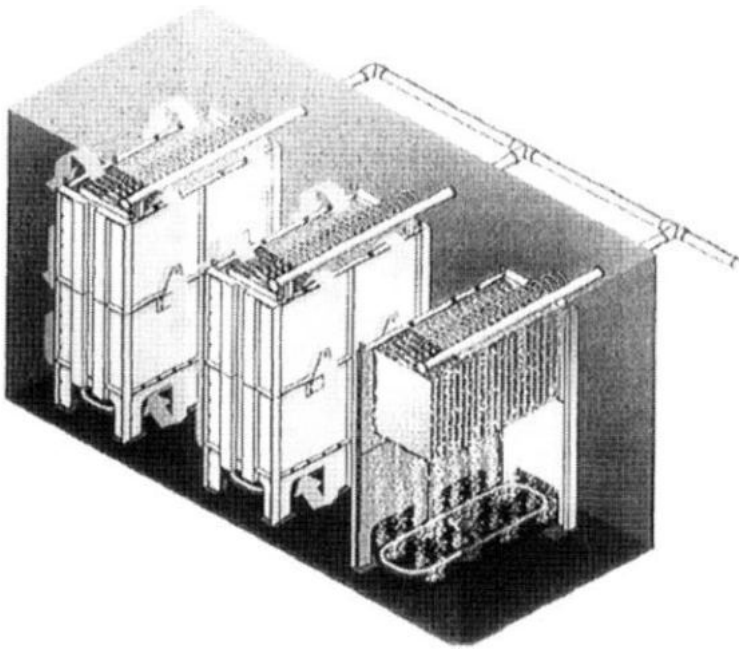
Figure 6.2. Schematic of the Kubota submerged membrane biroeactor. The arrows illustrate the flow of mixed liquor through the membrane module brought about by aeration from beneath the membrane plates. Also shown is the treated water collecting manifold to which each membrane module is attached.

6.1.2 Process Operation

During normal operation, the treated water flow through the membrane units, the permeate flux, is controlled by the liquid head above the membrane units; this gravity head is typically between 1 to 1.5 m. The number of units installed is dependent on the maximum flow rate required. Therefore the permeate flux determines the hydraulic retention time (see section 4.). Typically wastewater to be treated will pass through a degritter and a 2–3 mm perforated screen prior to the Kubota MBR (Figure 6.3). If complete nitrogen removal (nitrification/denitrification) is required, an anoxic zone with recycle from the aerobic activated sludge tank would receive the wastewater to be treated first.

Crude effluent feed

Degritting and screening
to 3 mm perforated

Gravity

| Optional recycle - return activated sludge | Gravity | Optional anoxic tank |

Gravity

To sludge treatment/storage

Pump

Aeration/Nitrification tank/s containing Kubota units

Gravity

Final effluent
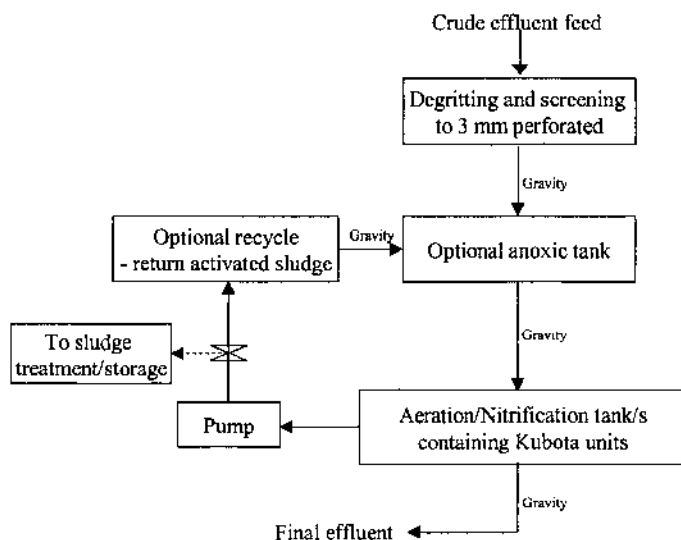
Figure 6.3. Typical flow diagram for the Kubota MBR

6.2 ZENON

Thetford Systems, Inc. of Michigan developed a sidestream MBR system, Cycle-Let, in the mid-1960s based on a two-stage single sludge aerobic/anoxic process. These systems used tubular ultrafiltration membrane units along with a two-pump feed and bleed loop. Process fluid from the aerobic reactor was pumped through a stationary screen, which was continuously cleaned by aeration. Permeate from the UF membranes was disinfected by UV and subsequently used for toilet flushing. Thetford Systems, Inc., which in 1994 became Zenon Municipal Systems, installed approximately 27 systems of this design between 1974 and 1982 for treating and recycling flushwater from small commercial facilities.

In the early 1980s, Thetford began applying this process to larger facilities such as major office buildings, shopping centres, industrial parks, sports facilities and other facilities where recycling flushwater was required to reduce wastewater discharge into the sewer.

The ZenoGem is the proprietary submerged MBR process that Zenon developed, and first commercialised for industrial applications using tubular ultrafiltration membranes in conjunction with suspended growth bioreactors. To

expand the application of this technology into large municipal wastewater treatment market, Zenon launched the development of hollow fibre membrane, named ZeeWeed. This product was first adapted to the commercial systems that were being operated or marketed by Zenon Municipal Systems

6.2.1 The ZeeWeed Membrane

ZeeWeed is a chlorine resistant microfiltration membrane with a nominal pore size of 0.1 μm. It is a hollow fibre with an outside diameter of 1.9 mm used in filtration from outside-in. A module consists of hollow fibres mounted on a vertical frame with permeate extraction from both ends. Coarse bubble aerators are integrated into the bottom header to gently agitate the hollow fibres and mix the contents of the tank.

Modules are assembled into cassettes, which constitute the physical unit immersed in the process tank. Several cassettes can be operated in parallel from a single pump. First generation ZW-150 modules have been superseded by ZW-500 modules, which more than double the membrane packing density and hence reduce the space requirements (Table 6.1).

Filtration is achieved at fluxes ranging between 40 and 70 $1 \text{ m}^{-2} \text{ h}^{-1}$ under a TMP of 10–50 kPa. The TMP is obtained from a combination of static pressure on the reactor side, and negative pressure on the permeate side using conventional centrifugal pumps. Periodic online backpulses with stored permeate are used to control fouling and to extend the interval between chemical cleanings. A key feature of the immersed membrane configuration is that the TMP is small and essentially uniform along the fibre length (neglecting small pressure losses on the lumen side), which leads to low fouling.

Table 6.1: The ZeeWeed membrane modules and cassettes

	ZW-150	ZW-500
Module dimensions (cm)	Height: 180 Width: 50 Thickness: 10	Height: 200 Width: 70 Thickness: 20
Filtration surface area (m²)	14	46
Number of modules per cassette	12	8
Cassette dimensions	Height: 200 Length: 190 Width: 70	Height: 200 Length: 180 Width: 70
Filtration surface area (m²)	168	368

6.2.2 The ZenoGem Process

The ZenoGem process can be operated with either the ZeeWeed membrane modules submerged directly into the bioreactor or in a separate tank to the bioreactor with recirculation of the reactor liquor to be filtered. When the membrane modules are submerged in the bioreactor only a single pump is necessary to provide a negative pressure for permeate extraction. This is the simplest configuration available and is used for large flow applications, such as municipal sewage works. There are several reasons for operating the process with the membrane external to the bioreactor; it is far easier to retrofit/upgrade an existing process (see case study in Chapter 7), operating the membrane tank under aerobic conditions and converting part of the bioreactor into an anoxic zone enables denitrification, and the membranes are easier to isolate for cleaning/inspection and maintenance.

Air to clean the ZeeWeed membranes is supplied by using coarse bubble aerators and normally satisfies approximately one half of the total biological oxygen demand. Fine bubble aeration is used to supply balance of oxygen. Most plants are designed for peak loads twice the average increasing the vacuum on the ZeeWeed membrane. Equalisation is used where peak loads exceed double.

End-suction centrifugal pumps are used for permeate suction, and self-priming pumps are not normally needed. Variable frequency drives are used for permeate pumps to meet variable flow requirements. Fouling management includes air scrubbing, in-situ maintenance cleaning with chlorine or other chemicals and scheduled recovery cleaning. Operating strategy focuses on reducing the frequency of recovery cleaning through proper operation and regular maintenance cleaning. Excellent effluent qualities can be consistently achieved at ZenoGem plants (Table 6.2).

Table 6.2: Typical municipal ZenoGem process effluent quality

BOD_5	< 2 mg l^{-1}
SS	< 1 mg l^{-1}
NH_3 N	< 0.1 mg l^{-1}
TN (cold climates)	< 10 mg l^{-1}
TN (warm climates)	< 3 mg l^{-1}
TP (chemical treatment)	< 0.1 mg l^{-1}
Turbidity	< 0.1 NTU
Total coliform	< 100 cfu/100 ml
Faecal coliform	< 20 cfu/100 ml
SDI	< 3

Total biological sludge generation varies between 0.02 kg kg^{-1} COD for industrial plants to 0.4 kg kg^{-1} COD for municipal facilities. This depends on

factors such as the HRT, MLSS concentration and SRT and coagulant addition for phosphorus removal.

6.3 ORELIS & MUTSUI CHEMICALS

Orelis is a subsidiary of the Rhodia Group. It has sold the license of its ultrafiltration plate and frame systems (Pleiade) to the Japanese Company Mitsui Chemicals Inc (MCI). The Pleiade membrane module has been employed in an aerobic MBR process developed by Orelis and MCI in which the membrane is external (i.e. sidestream) to the bioreactor. The flat-sheet membrane made from acrylonitrile co-polymers has a MWCO of 40,000 Da. These membrane flat sheets are held in a stainless steel housing external to the bioreactor (Figure 6.4). The liquid velocity through the membrane module, as determined by the recirculation rate, ranges from 1 to 2 m s^{-1} depending on the filtrability of the bulk liquor. Air, to provide mixing in the bulk liquid and oxygen for aerobic biodegradation, is entrained into the recirculation loop via a venturi (Figure 6.5). The energy requirement of the process ranges from 1 to 3 kW m^{-3}. Permeate flux rates of approximately 100 l m^{-2} h^{-1} can be maintained for 30–45 d without membrane cleaning.
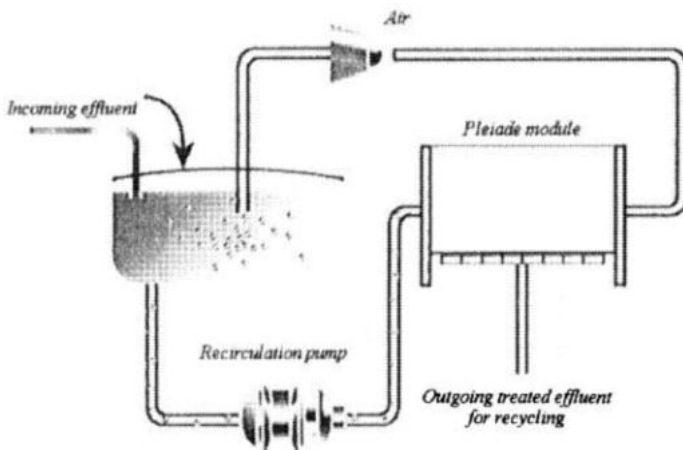
Figure 6.4. Schematic of the Pleiade aerobic membrane bioreactor, with membrane unit external to the activated sludge reactor.

6.3.1 Ubis

Since 1980, Mitsui Chemicals Inc. has been using the previous experience of the Rhodia Group to develop a system for in-building wastewater treatment, with its first plant in Tokyo in the Marunouchi Building in 1981.

The Ubis system enables the recycling of wastewater from kitchens, wash basins and toilets; collectively known as greywater. The wastewater is transferred to an activated sludge bioreactor which has a high sludge concentration (approximately 20 g l^{-1}) and a high degree of mixing. As a consequence of these two factors, and the separation of the HRT from the SRT that is common to all MBR processes, the HRT is reduced to approximately 1 h. This compares with 24–48 h HRT necessary in conventional wastewater treatment systems treating greywater.

As will other MBR process the water passing through the membrane is free of suspended solids, viruses and bacteria. Treated water is stored in a buffer tank where a small amount of sodium hypochlorite is added. It is then reused for flushing toilets and a volume equal to the quantity of potable water legally required in a building (kitchens, basins, showers) is put back into the urban wastewater network. M.C.I. has adapted the Ultra Biological System (Ubis) developed by Orelis as the basis for the Activated Sludge and Membrane Complex system (Asmex) used for the treatment of highly concentrated effluents.

6.4 MEMBRATEK, WEIR ENVIG, AQUATECH AND BIOSCAN A/S

Weir EnVig (South Africa), AquaTech (Korea) and Bioscan A/S (Denmark) all employ Membratek (South Africa) tubular membranes in their sidestream MBR processes. Membratek developed the Anaerobic Digestion Ultra-Filtration (Aduf) in the late 1980s for the treatment of high strength industrial effluents, principally food processing and brewery production effluents. The Biorek (Biological Recirculation and Kraft) process, developed by Bioscan A/S, has an Aduf process coupled to an ammonia stripper and reverse osmosis unit for the treatment of agricultural waste. AquaTech uses Membratek Memtuf membranes in its aerobic MBR process, Biosuf, used principally for municipal wastewater treatment.

The Membratek tubular polyethersulphone ultrafiltration membrane has a pore size typically 0.10 μm and the MWCO varies from 6,000 to 80,000 Da. The length of each tube is 3 m and each module has two parallel flow paths of 20 series tubes each. It is claimed that these unsupported tubular modules are economically competitive with conventional tubular systems as they do not

require the customary high-pressure support structure. The Memtuf membrane module used in AquaTech's Biosuf aerobic MBR process has a MWCO of 40,000 Da, an inside diameter of 8.9 mm, an operating inlet pressure of 4.5–5.5 kg cm^{-2} and an average flux of 40 lm^{-2} h^{-1}.

The sidestream (i.e. external) compact ultra-filtration modules provided by Membratek can be retrofitted to conventional anaerobic wastewater treatment processes. The Aduf process was introduced in the late 1980's and early 1990s in South Africa from demonstrations units (0.05–3.0 m^3) to commercial processes (80 to 2610 m^3).

The Biorek process developed by Bioscan A/S treats agricultural slurry by coupling an Aduf, an ammonia stripper and a reverse osmosis (RO) unit. The RO process is capable of purifying the product from the stripper unit to potable water. In the Aduf process incorporated in the Biorek treatment system the MWCO of the Membratek membranes is 40,000 Da, which ensures that macromolecules and bacteria are rejected by the membrane. Tubular membranes with an internal diameter of 12.5 mm are housed in low cost aluminium modules.

6.5 WEHRLE WERK AG

6.5.1 The Biomembrat process description

Biomembrat is an aerobic sidestream MBR originally developed at the University of Stuttgart, and further optimised and applied by Wehrle-Werk AG. In this process the airtight bioreactor is subject to pressures of up to 3 bar; this being achieved by air and/or oxygen supply. Higher oxygen solubility at the operating pressure results in an improved supply and utilisation of oxygen. This has particular benefits when treating high strength industrial wastewaters. The reduced air/gas discharge reduces the stripping of volatile pollutants into the atmosphere and odour emissions. Volumetric loads can be regulated by the modular construction and operation of the membrane filtration modules, as well as by changes in the process parameters.

Biomembrat plants treating landfill leachate, with typical flows of 100 m^3 d^{-1} rising to a maximum of 600 m^3 d^{-1} and requiring an aeration volume of between 50 m^3 and 250 m^3 are operated under pressure. Larger plants treating either chemical and food industry wastewaters with typical flows up to 2,500 m^3 d^{-1} require bigger aeration volumes of around 1000 m^3 that are not operated under pressure.

The non-pressurised Biomembrat plants operate at around 20 kg MLSS m^{-3}, while the pressurised plants operate in the range of 20–30 kg MLSS m^{-3}. The retained biomass is continuously recirculated to the reactor by ultrafiltration.

Typically a polysulfon based membrane has been used in the treatment of landfill leachate by the Biomembrat process. The ultrafiltration unit, which is external to the bioreactor, usually consists of several loops in parallel, each having up to six modules in series. Each module contains several tubular membranes with a diameter ranging from 5 to 12 mm. The number of membrane tubes varies from 9 to >300 with a surface area per module of 0.9–23 m^2. The crossflow velocity and flux varies from 2.5 to 4.5 m s^{-1} and 50 to 250 l m $^{-2}$ h^{-1}, respectively, depending on the type of wastewater. The lower flux rates occur when treating high strength industrial wastewaters at a high loading rate.

Crossflow operation enables in-situ cleaning with either water or a chemical cleaning agent. As the membrane unit is external to the bioreactor and designed in loops, it is relatively easy to isolate individual loops and carry out a clean procedure in the same direction as normally travelled by the bioreactors bulk liquid. Wehrle Werk AG claim that this method of cleaning is much more efficient than the backwashing procedure typical of MBR processes that have submerged membrane units, particularly with regard to removing precipitates. Such a maintenance clean is typically required once or twice a year. The life expectancy of the membrane modules, is in the range of 4–8 years.

6.5.2 Process Performance

As with all commercial MBR process the Biomembrat can achieve a high degree of organic removal at high loading rates and influent concentrations. Almost complete COD removal at influent concentrations of up to 15,000 mg l^{-1} has been demonstrated and influent concentration as high as 100,000 mg l^{-1} are feasible. The HRT depends on the wastewater type; easily biodegradable wastewater requires a minimum of 8 h, whereas difficult to degrade industrial effluents can take up to 24 h.

As with other MBR process excess sludge production in the Biomembrat process can be lower than the conventional activated sludge process; pilot trials treating dairy effluents have demonstrated close to zero excess sludge production (Table 6.3).

Table 6.3: The correlation between F:M ratio, volume loading and excess sludge production in the Biomembrat process

F:M ratio (kgBOD kg MLSS^{-1} d^{-1})	Volume loading (kgBOD m^{-3} d^{-1})	Yield (kgMLSS kgBOD^{-1})	sludge age (d)	Remarks
0.09	1.8	0.09	120	Low excess sludge
0.5	10	0.5	5	Low space Requirement

For nitrogen removal, plant design is based on a nitrate sludge loading of between 0.03 and 0.1 kg N kg TS^{-1} d^{-1}. At influent ammonia concentration of up to 2000 mg l^{-1} greater than 90% denitrification can be achieved. Successful nitrification/denitrification in the Biomembrat process has achieved at ammoniacal-nitrogen concentrations as high as 12,000 mg l^{-1}. The use of an additional carbon source, usually methanol or acetic acid, has been used because to the nature of the wastewater. Phosphorus removal does naturally occur in the Biomembrat process but can be enhanced by adding ferric chloride.

6.5.3 The Biomembrat-Loop Process

The Biomembrat-Loop process is an aerobic sidestream MBR (Figure 5.6). Unlike most other sidestream MBR processes air is supplied to the vertically mounted ultrafiltration tubular membrane module as well as to the bioreactor. In this MBR process the recirculation pump is only used during peak volumetric load to increase the flux rate. Therefore, during normal operation low energy operation is achieved by using airlift in the membranes module. In normal airlift operation of the membrane process, with low cross-flow velocity and low energy requirements, flux rates of 20–40 l m^{-2} h^{-1} can be achieved. The treated water is removed under a vacuum of no more than 0.5 bar.
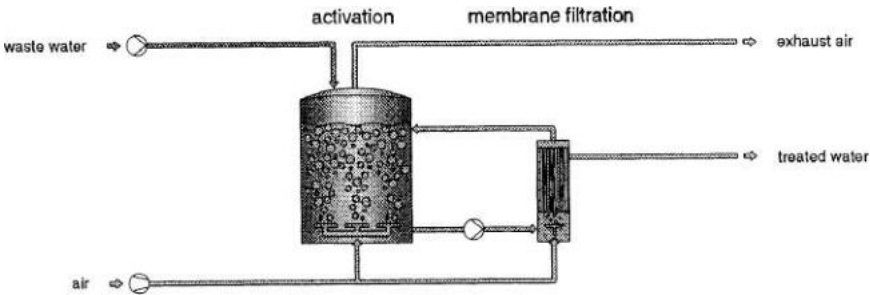
Figure 6.5. Schematic of the Biomembrat-loop

The increasing specific energy consumption as the flux rate increases can be divided into the three different modes of operation; air injection, a combination of air injection and recirculation pump and solely recirculation pump (Figure 6.5). The required specific energy in standard operation with air injection is between 0.5 and 1.5 kWh m^{-3} permeate. If higher permeate fluxes are required an energy consumption of 1.5 to 3.5 kWh m^{-3} can be expected due to the need to operate the recirculation pump. The energy requirement can be divided into hydraulic and biological demands. As a rule of thumb, and excluding nitrogen removal, the following formula can be applied.

$$\text{Energy Required} = 1.6 \text{ kWh kgCOD}^{-1} + 2.6 \text{ kWh m}^{-3}$$
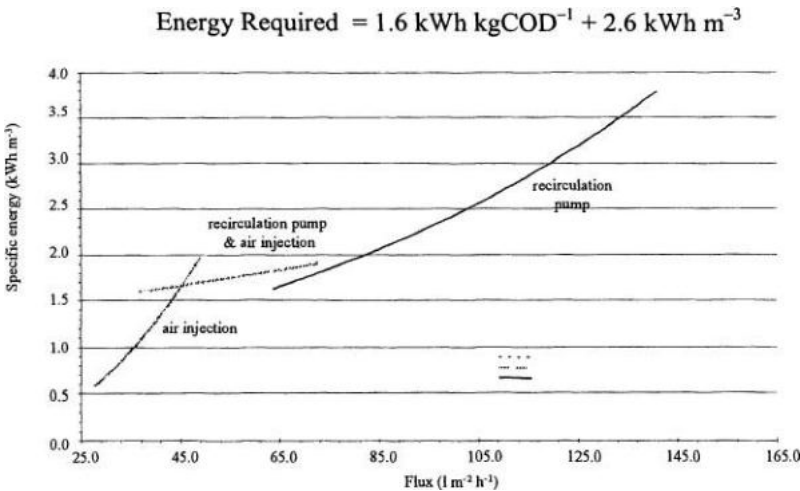
Figure 6.6. Specific energy consumption of the Biomembrat-loop

6.6 US FILTER

US Filter are part of the Vivendi Group, which also includes the Generale de Eaux and OTV. Both USF Memcor and USF Gütling, like OTV, have in the past developed their own MBR processes.

Memcor developed the Membio process, a sidestream MBR, in Australia towards the end of the 1980s. Hollow fibre polypropylene membranes with a 0.2 μm pore size are operated in an out-to-in mode. Accumulated solids are removed from the outer surface of the hollow fibres by periodic high pressure air injection into the fibre lumen. Air passes through the membrane wall and dislodges accumulated sediment (Gorden-Werner *et al.*, 1993).

The Biosep process developed by OTV is essentially the same as Zenon's ZenoGem process, utilising ZeeWeed hollow fibre membrane cassettes submerged directly into the bioreactor tank.

More recently the Germany based company USF Gütling have developed the Kompajet, a sidestream membrane bioreactor process that uses tubular membranes (Table 6.4). Like the Biomembrat-loop process either airlift or a recirculation pump can be used to provide the necessary crossflow at the membrane surface. The liquid crossflow velocity increases from 1.5 to 3.5 m s^{-1} as the air flow rate is increased from approximately 5–40 m^3 h^{-1}. Permeate fluxes of 20–50 l m^{-2} h^{-1} can be achieved using airlift to provide crossflow with an energy requirement of 1.0 to 2.5 kWh m^{-3}. The TMP is aided by a vacuum on the permeate side of the membrane. Switching to the recirculation pump increases the performance and energy requirement of process; at a crossflow velocity of 3.0 to 6.0 m s^{-1} permeate fluxes of 80 to 140 l m^{-2} h^{-1} require between 5.0 and 6.5 kWh m^{-3} of energy.

Table 6.4. Characteristics of the membrane used in the Kompajet MBR.

Characteristic	
MWCO (D)	100,000
Membrane material	Polyethersulfone or polyetherketone
Support material	Polypropylene-polyester
Diameter (mm)	8–11
Surface area per membrane module (m^2)	2–12

6.7 DEGREMONT

The process developed at CIRSEE, the research arm of Lyonnaise des Eaux, and commercialised by Degremont is the only commercial MBR process to use ceramic membranes. Though more expensive, ceramic membranes have a much higher tolerance to extremes of pH (0 to 14), pressure (<10 bar) and temperature (<350 °C) than the organic polymers used in other MBR processes. The ceramic membranes are situated externally from the bioreactor with the recirculation pump providing the high pressures necessary. A typical flux of $150 \ l \ m^{-2} h^{-1}$ can be maintained at TMPs of 0.8–1.2 bar over approximately a 20 d period after which an intensive and efficient chemical cleaning is needed to restore membrane performance. Due to the high flux rates capable when using ceramic membranes, the process has been applied to the treatment of high strength industrial wastewaters as the relative energy consumption used for filtration is reduced and thus the process is claimed to be competitive with other MBRs.

6.8 REFERENCES

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7

Membrane bioreactor case studies

7.1 KUBOTA, MBR TECHNOLOGY

The Kubota MBR was first utilised in a commercial treatment plant in 1990 and there are over 200 plants installed worldwide (Table 7.1).

Table 7.1: Applications of the Kubota MBR (Courtesy of the Kubota Corp., January, 1998)

Wastewater Type	# of Plants	Capacity Range	# of Membrane Cartridges	Year
In-building	23	16–300	50–3,600	1991–1999
Municipal	4	1–110	50-625	1997–1998
Domestic	65	10-800	7–3,600	1990–1997
Industrial	50	4–1,000	7–3,600	1995–1997

Over the past five years there has been a dramatic increase in both the number of operational Kubota MBR plants and their scale (Table 7.2.). In the United Kingdom, the Kubota MBR is being applied to domestic wastewaters with flows in excess of 10,000 m^3 d^{-1}. Commercial submerged membrane

bioreactor systems have now been operational for almost a decade and have proven both reliable and simple to operate.

Table 7.2: Number of Kubota MBR plants worldwide (Courtesy of MBR Technology, 1999)

Year	No of plants	Largest plant ($m^3 d^{-1}$)
1993	4	125
1995	20	250
1997	70	800
1998	150	1907
1999	237*	7100
2000	?	12,700*

*Under construction.

Since 1992 the design flux rate, governed by the hydrostatic head above the membranes, has been doubled and thus reduced plant size and power consumption. Furthermore, the projected membrane life has increased from three to eight years, the scale up of manufacture has reduced membrane fabrication costs and plant design and operation has been simplified reducing capital costs and maintenance requirements in operation.

7.1.1. Case study: municipal wastewater treatment

Following successful pilot-trials of the Kubota MBR process, MBR Technology built their first commercial plant at Porlock on the North Somerset coast in England in 1997 (Churchouse, 1999; Churchouse and Wildgoose, 1999). Porlock is a village with approximately 4000 people within the catchment area. The site is overlooked by both the village and surrounding hills and the effluent discharges into the sea near a bathing beach. The requirement was thus for a high quality compact treatment process that could blend into the surroundings. Planning requirements dictated that the plant should be housed within a local stone faced building to fit in with the adjacent farm property.

Operation was started in February 1998 and the plant has a maximum treatment capacity of 1900 m^3 d^{-1}. A total of 24 Kubota membrane units are contained in four aeration compartments with the treated effluent removed under gravity through the wall of the tank (Figure 7.1). Owing to the requirement for tidal storage, pumps are used to lift the treated effluent into the tidal tank.
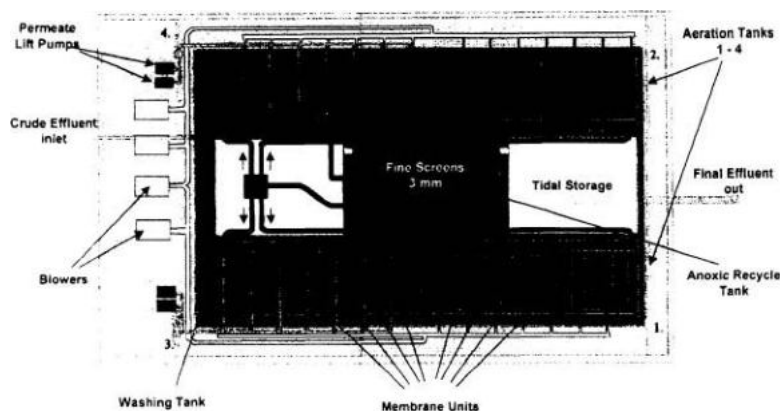
Figure 7.1. Schematic layout of the Porlock sewage treatment plant (Courtesy of MBR Technology, UK).

During the first 14 months of operation, when exhaustive analysis was done, the effluent quality was excellent, with the performance matching that from the original pilot-plant trials (Table 7.3). Final effluent BOD did not exceed 5 mg l^{-1} and was not influenced by variations in influent BOD. Bacterial disinfection averaged 6 log removal with virus removal averaging a 4 log for both enterovirus and coliphage. Treated permeate turbidity on-line averaged 0.3 NTU.

Table 7.3:. Wastewater characteristics and treatment performance of Kubota MBR at orlock, North Somerset, England (02/98-04/99).

Pollutant	No. of samples	Influent Range	Influent average	Effluent average	% Removal
Suspended	56	<30–800	230		>99.5
solids (mg l^{-1})	256			<1	
Turbidity (NTU)	On-line	>100	-	<0.4	
BOD	61	<30–650	224		>97.2
(mgO$_2$ l^{-1})	264			<4.0	
Faecal coliforms (10^6 100 ml^{-1})	55 252	0.9–64	10.1	<0.00002	>99.9998 (>log 5.7)
Faecal streptococcus (10^6 100 ml^{-1})	55 252	0.1–30	1.32	<0.00001	>99.9993
Coliphage virus (10^6 100 ml^{-1})	28 125	<29–6320	811	<0.19	>99.98

Installation and commissioning was straightforward with membrane units tested with clean water before start-up. However, some problems were identified during the commissioning of the plant and were subsequently rectified. These were principally associated with peripheral mechanical and engineering equipment or the sewerage distribution system.

Tidal related seawater intrusion (up to 70% of incoming flow at times). Prevented by replacing a small section of badly damaged sewer near the beach.

Poor upstream fine screen performance resulted in a change from 2 mm wedgewire to 3 mm perforated screens. Uneven flow distribution from the flow-split chamber to the 4 aeration tanks was rectified and modifications to the software control system to improve the effectiveness of a water flushing system for the aeration diffusers were made.

No membrane failures have so far been observed out of the 3600 panels installed. Maintenance of the membrane components has thus far been limited to the occasional water flushing through the aeration diffusers, and the cleaning of flow-meters and turbidity flow cells. An *in situ* chemical clean with a backwash of 0.5% sodium hypochlorite was carried out after approximately 9 months' operation. This required approximately 5 h downtime for each of the four aeration tanks. Chemical costs were negligible. Used cleaning chemicals were removed by the permeate manifold and returned to the works inlet. This ensured free chlorine levels were below 0.2 mg l^{-1} before the tank was returned to normal operation. Measurements of trihalomethane (THM) levels indicated a maximum hourly level of 0.052 mg l^{-1} chloroform in the final effluent immediately following cleaning. Chloroform levels reduced to 0.002 mg l^{-1} within 24 h.

Foaming was observed on one occasion owing to an incident where a large quantity of scented surfactant appeared in the wastewater. The foaming was rapidly controlled by allowing the foam to overflow to the central anoxic tank where a single spray was set up. No other foaming or sludge bulking has been observed. No significant odours were apparent in the treatment building, with hydrogen sulphide levels of <0.002 mg l^{-1} measured. No odour control equipment has been installed in the treatment building. Sludge ages for the plant have generally been in the range of 30–60 d depending on season. Sludge production has been measured in the range of 0.38–0.5 kg kgBOD^{-1}. This equated to between 3 and 6.5 m^3 per day of 2% sludge at Porlock dependent on season.

7.2 ZENON

The ZenoGem submerged MBR process has provided in-building or on-site treatment of wastewaters from residential, commercial and industrial sources with flows ranging from 10 to 200 m^3 d^{-1} since 1994 (Table 7.4). During this period ZeeWeed based ZenoGem MBRs, with capacities of 100–500 m^{-3} d^{-1}, have been used for the treatment of landfill leachates in The Netherlands (Husain and Cote, 1999). Many of the in-building ZenoGem processes, treating either greywater or blackwater, enable the reuse of the treated water for toilet/urinal flushing and landscape irrigation and thus reduce the wastewater discharge to sewer. The process is also being used for municipal wastewater treatment (Table 7.5).

Table 7.4. ZenoGem installations in the 10–200 m^3 d^{-1} range (February 1999, courtesy of Zenon Environmental Inc., Canada)

Type	Number
Residential	10
Office	15
Shopping Centres	10
Schools	6
Industrial	5
Hotels and Resorts	6

Table 7.5. Municipal ZenoGem installations (February 1999, courtesy of Zenon Environmental Inc., Canada)

Installation	Capacity ($m^3 d^{-1}$) Normal	Peak	Commissioned
Germany	1344	3240	Spring 1999
France	900		Summer 1999
Florida	1300	3000	Spring 1999
Florida	1900	3800	Spring 1999
Egypt	3800	7600	Spring 1999
Arizona	1000	2000	Spring 1999
Egypt	1900	3800	Autumn 1998
British Columbia	3800	7600	Summer 1998
Colorado	3800	5700	Summer 1998
New Jersey	680	860	Spring 1998
Ontario	1000	2000	Summer 1997
British Columbia	380		Autumn 1996
British Columbia	1130		Autumn 1996

7.2.1 Case study: retrofit of sequencing batch reactor

In this case study an existing municipal wastewater treatment plant was successfully converted to the ZenoGem MBR process in order to improve effluent quality and increase flow capacity. Effluent total nitrogen and total phosphorus requirements for this plant were stringent. Arapahoe County Water and Wastewater Authority's Lone Tree Wastewater Treatment Plant (Englewood, Colorado, USA) required an expansion of its 3030 m^3 d^{-1} treatment plant. The plant consisted of two SBRs in square concrete tanks. Each tank had an operating volume of 1135 m^3. The capacity of the plant needed immediate upgrading to 3785 m^3 d^{-1} and effluent quality needed to be improved to achieve limits for phosphorus (0.2 mg l^{-1}) and total nitrogen (10 mg l^{-1}). The following alternatives were considered by the county and its engineers: (1) expand the existing SBR and add advanced treatment for enhanced nutrient removal; (2) transform the SBR tanks into an advanced activated sludge plant by adding clarifiers and sand filters or; transform one SBR tank into a ZenoGem MBR. The county anticipated that the plant would need to be expanded beyond (1) in the near future and land availability was of prime consideration in the process selection. As a result the ZenoGem technology was selected on the basis of its capital and operational cost as well as the high effluent quality, suitable for water reuse.

7.2.1.1. Plant Design

For the initial expansion, only one of the two existing SBR tanks was used for the ZenoGem process. The second tank is being used as a reserve for the next stage of expansion. The existing coarse bar screen was upgraded to a fine 3 mm screen. The existing jet aeration system was replaced with a fine bubble diffuser grid and existing blowers were used for all air requirements.

A concrete baffle was constructed in the first tank separating it into a plug-flow, anoxic zone preceding a completely mixed, aerobic zone aerated with the fine bubble diffuser grid. One hundred and forty-four ZW-500 membrane modules were contained within two steel frames located within the aerobic zone. Nitrified mixed liquor was pumped from the aerobic zone to the anoxic zone at a rate equivalent to six times the influent wastewater flow rate. Phosphorus removal is achieved through 50 mg l^{-1} ferric chloride addition to the screened wastewater, prior to entering the anoxic zone.

The aerobic zone provided a working volume of 606 m^3, representing a nominal HRT of 3.8 h at design flow. The anoxic zone provided a working volume of 379 m^3, representing a nominal HRT of 2.4 h at design flow (Table 7.6).

Table 7.6: Process operating conditions for the Lone Tree ZenoGem retrofit

Design Average Flow	3785 m^3 d^{-1}
Design Peak Flow	5678 m^3 d^{-1}
Anoxic Working Volume	379 m^3
Aerobic Working Volume	606m^3
MLSS Range	12,000–15,000 mg l^{-1}
MLVSS Range	8,000–10,000 mg l^{-1}
Solids Retention Time	15–20 d
Aerobic Hydraulic Retention Time	3.8 h @ design flow
Anoxic Hydraulic Retention Time	2.4 h @ design flow

The retrofit was completed in July 1998, and the plant seeded in August 1998.

7.2.1.2. Influent Wastewater Characteristics

The influent wastewater characteristics for the first 9 months of operation were monitored (Table 7.7). The pollutant concentrations observed were typical for raw sewage in the region and were consistent with historical values observed at the plant. The wastewater temperature remained relatively stable throughout the year, as well water served as the primary water source for the community, and the county was served by a separate sewer system minimising rain water and snowmelt infiltration. The influent wastewater temperature throughout the time period reported ranged from 13 to 16 °C.

Table 7.7: Monthly Average Influent Wastewater Characteristics

Month	Flow (m^3 d^{-1})	Carbonaceous BOD (mg l^{-1})	NH$_3$-N (mg l^{-1})	Total phosphorus (mg l^{-1})	Total suspended solids (mg l^{-1})
August[a]	3400	183	30.2	13.8	253
September	3200	187	33.0	10.8	224
October	3200	178	35.2	12.8	251
November	3180	221	38.1	Not available	262
December	3940	185	34.2	Not available	230
January	3104	237	34.0	12.4	282
February	3104	233	34.4	16.1	262
March	3180	217	37.5	17.1	237
April	3290	184	37.2	21.6	272
Average[b]	3290	205	34.6	15.8	251

[a] Sampling began 14 August 1998

[b] Total samples: 32

7.2.1.3. Sludge Yield

The ZenoGem plant was operated at an MLSS concentration of 12,000–20,000 mg l^{-1}. The volatile fraction of the mixed liquor solids ranged from 55% to 70%. This low volatile fraction resulted from the relatively high ferric chloride dosage required to achieve the low effluent phosphorus concentrations. A waste sludge flow rate between 53 m^3 d^{-1} and 68 m^{-3} d^{-1} was used to maintain the target MLSS concentration. The resulting sludge age was 14.4–18.6 d. The net yield was estimated at 0.8 kg TSS kg BOD^{-1} applied, of which 25% was chemical sludge associated with ferric chloride addition.

7.2.1.4. Performance

Effluent quality consistently complied with the criteria set by the county in terms of carbonaceous BOD (<5 mg l^{-1}), total suspended solids (<5 mg l^{-1}), total phosphorus (0.2 mg l^{-1}), and total nitrogen (<10 mg l^{-1}) (Table 7.8; Figures 7.2–7.5). Total nitrogen concentrations were slightly elevated for the first 4–6 weeks of operation as the system was seeded with a non-nitrifying sludge. As soon as nitrification was achieved, total nitrogen concentrations in the effluent remained consistently below 6 mg l^{-1}.

Table 7.8: Average monthly effluent wastewater characteristics from the ZenoGem at Lone Tree WWTP.

Month	Carbonaceous BOD (mg l^{-1})	Total N (mg l^{-1})	Total P (mg l^{-1})	Total Suspended Solids (mg l^{-1})
Aug	3.5	17.2[b]	0.11	2.1
Sept	3.1	10.2[b]	0.12	1.8
Oct	1.2	4.2	0.13	2.0
Nov	1.9	4.2	0.08	2.0
Dec	1.8	3.5	0.16	2.9
Jan	3.2	4.6	0.07	2.1
Feb	2.4	4.1	0.05	2.2
Mar	2.2	5.6	0.07	2.5
Apr	3.0	5.1	0.13	2.3
Average[a]	2.8	4.4[b]	0.11	2.2
Criteria	<5	<10	0.2	<5

[a] Total samples: 120.

[b] During the first six weeks of operation the sludge was being conditioned for optimal nitrification; these numbers are not included in the overall average.
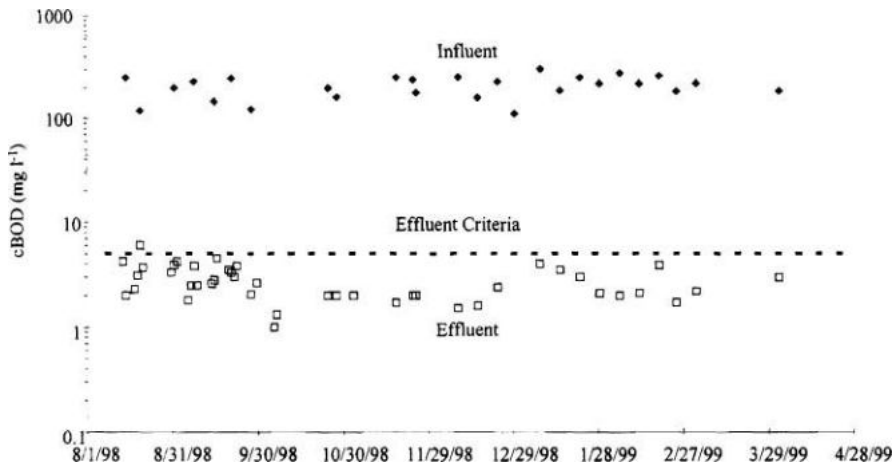
Figure 7.2. Influent, effluent and criteria concentrations of carbonaceous BOD at the ZenoGem MBR at Lone Tree WWTP.
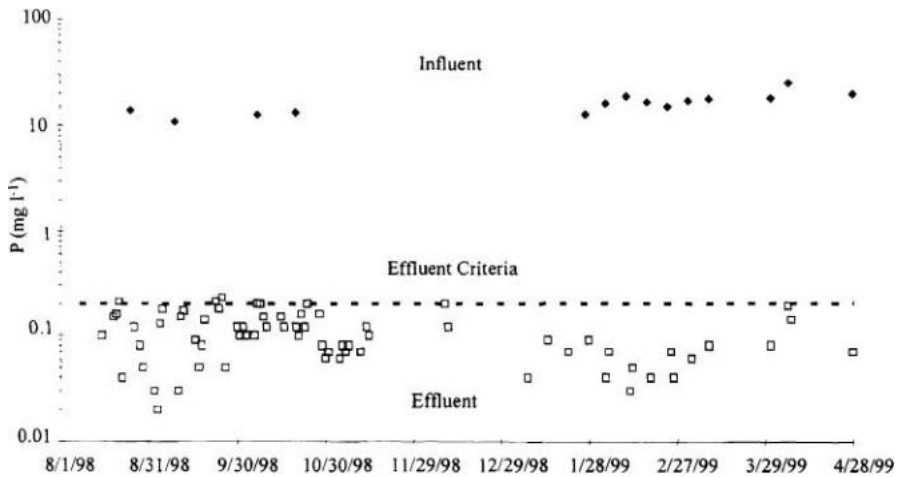
Figure 7.3. Influent, effluent and criteria concentrations of total phosphorus at the ZenoGem MBR at Lone Tree WWTP.
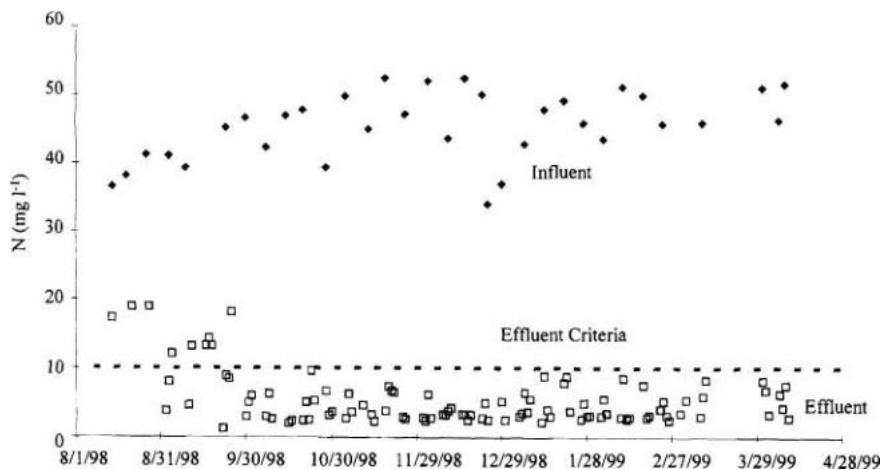
Figure 7.4. Influent, effluent and criteria concentrations of total nitrogen at the ZenoGem MBR at Lone Tree WWTP.
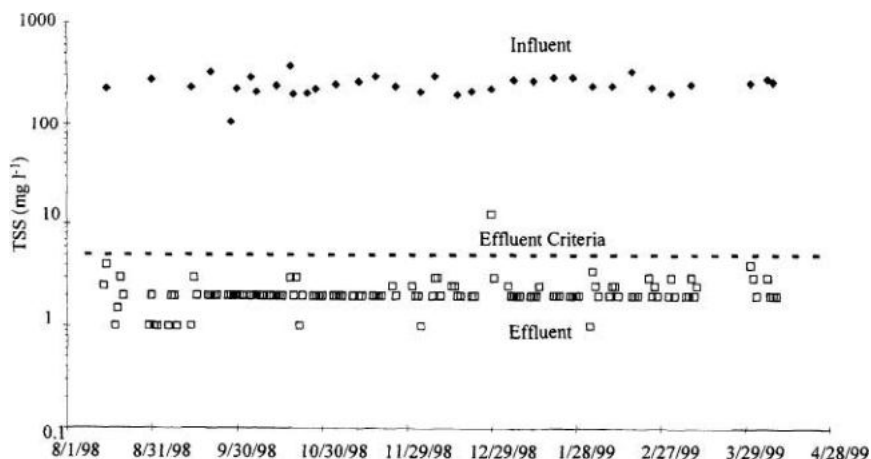
Figure 7.5. Influent, effluent and criteria concentration for total suspended solids at the ZenoGem MBR at Lone Tree WWTP.

7.3 ORELIS

The process has been trialled in France for the treatment of wastewaters from hospitals and urban power stations since the mid-1970s. The Pleiade MBR has been incorporated into two systems for the treatment of two different wastewater types: the Ubis system has been used for the in-building treatment of greywater for water reuse, where the compactness of the system is the main advantage; and the Asmex system has been used for the treatment of highly polluted industrial effluents, where the efficiency of the MBR is the key advantage.

By 1999 there were approximately 125 commercial aerobic MBRs using Pleiade membrane modules in operation worldwide, with 15 in Europe (Table 7.9) and the remainder in Japan.

Table 7.9: Orelis membrane bioreactors in Europe

Wastewater Type	Location	Capacity $(m^3 d^{-1})$	#	Commissioned
Cosmetics	France	250	1	1994
Leachate	Europe	20–200	7	1994–1998
Urban Effluents	France	120		1996
Dairy	France	50–120	3	1998-under construction
Truck Washing Station	Belgium	100	1	1998
Abattoir	France	350–1000	2	1999-under construction
Textile	Portugal	50	1	1999

In Europe most of Orelis systems are dedicated to industrial wastewater treatment. In Japan the Pleiade MBR, is mainly used for domestic wastewater treatment and recycling. More than 45 Ubis systems are used in Japanese buildings and large hotels.

7.3.1 Case Study: in-building greywater treatment and recycling

A 500 $m^3 d^{-1}$ Ubis plant has been installed in the basement of the Mori Building in Tokyo, Japan (Figure 7.6). To treat a 100 $m^3 d^{-1}$, a plant requires a 45 m^3 footprint, a membrane module with 34 m^2 of membrane surface area and a 6 m^3 bioreactor. The space saving for a 100 $m^3 d^{-1}$ Ubis process compared with a conventional treatment system is equivalent to 25 car parking places. The design

of an ultrafiltration module using turbulent generator allows very high permeate fluxes to be maintained over long periods. Over a 45 d period the flux will decrease from 120 to 100 l m^{-2} h^{-1}. *In situ* chemical cleaning is performed every 45 d and electrical consumption is approximately 3 kWh m^{-3} of treated water. The Mori Building plant performs well and the effluent is good enough for reuse (Table 7.10).

Table 7.10: Performance of Ubis Membrane Bioreactor in the basement of the Mori Building, Tokyo, Japan. This system as commissioned in 1986 with a design capacity of 500 m^{3} d^{-1} (Courtesy of Mitsui Chemicals Inc., Japan)

	Influent			Treated water for reuse		
	Average	Max.	Min.	Average	Max.	Min.
Temperature (°C)	19.5	22.5	15.5	26.3	30.5	21.0
Ph	6.6	8.5	5.7	6.8	7.9	6.1
COD (mg l^{-1})	89.1	140	12	12	21	<5
BOD (mg l^{-1})	349	577	120	3.7	5.5	1.8
SS (mg l^{-1})	96.5	160	33	b.d.	b.d.	b.d.
n-hexane extract (mg l^{-1})	11.7	12.4	11	<1	<1	<1
MBAS (mg l^{-1})	6.5	9.9	3.1	0.3	0.4	0.2
Colon bacilli (cfu 100 m l^{-1})	19×10^{4}	22×10^{4}	15×10^{4}	b.d.	b.d.	b.d.

b.d., below detection.
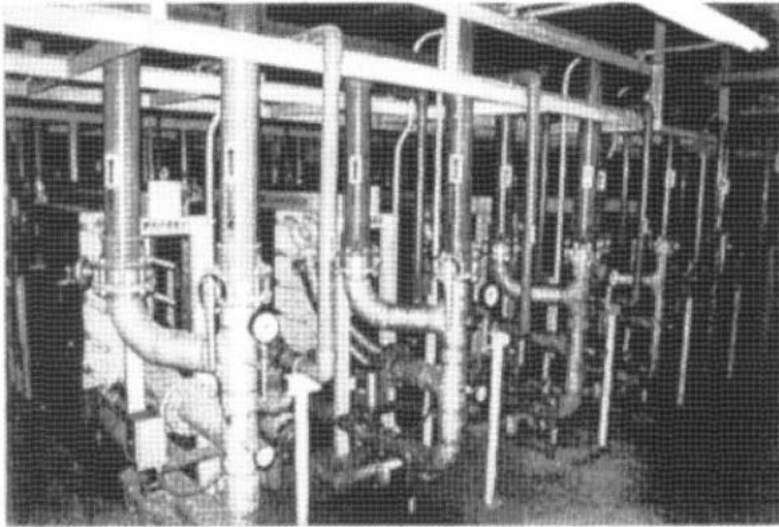
Figure 7.6. A Ubis Membrane Bioreactor in the basement of the Mori Building, Tokyo, Japan. This system as commissioned in 1986 with a design capacity of 500 m^3 d^{-1} (Courtesy of Mitsui Chemicals Inc., Japan).

7.4 MEMBRATEK, WEIR ENVIG, AQUATECH AND BIOSCAN A/S

7.4.1 The Biosuf process by AquaTech

The Memtuf membrane module used in AquaTech's Biosuf aerobic MBR process has a MWCO of 40,000 Da, an inside diameter of 8.9 mm, an operating inlet pressure of 4.5–5.5 kg cm^{-2} and an average flux of 40 l m^{-2} h^{-1}. By the end of 1999 there were 20 AquaTech Biosuf processes in operation in Korea, with another two to be commissioned during 2000 (Table 7.11). Most of these processes were used for the treatment of night-soil, agricultural wastewaters or domestic wastewaters. Biosuf processes that treat either night-soil or a combination of night-soil and livestock wastewaters achieved high percentage dissolved organic, suspended solid and nitrogen removal. With influent total nitrogen concentrations averaging 4500 mg l^{-1} about 99% was removed from

the treated water. The processes at Chilgok, Kunwee, Chiak, Pyungchang and Pyungchan, all of which are situated at highway services stations incorporate some form of reuse of the treated water.

Table 7.11: Biosuf processes installed by AquaTech in Korea

Location	Type of waste	Capacity $(m^3 d^{-1})$	Influent BOD $(mg\ l^{-1})$	SS $(mg\ l^{-1})$	Effluent BOD $(mg\ l^{-1})$	SS $(mg\ l^{-1})$	Start date
Ichon City	Night-soil Livestock	150	20,000	28,000	<5	b.d.	11.96
Paju City	Night-soil	60	20,000	25,000	<5	b.d.	12.97
Paju City	Night-soil	80	20,000	25,000	-	-	04.99
Gohung	Night-soil Livestock	95	20,000	28,000	-	-	07.99
Haman	Night-soil	25	12,000	24,000	-	-	09.99
Imsil	Night-soil Livestock	130	26,000	33,000	-	-	10.99
Paju City	Live-stock	200	-	-	-	-	05.00
Chilgok	Domestic	100	200	170	<1	b.d.	02.97
Kunwee	Domestic	100	220	150	<1	b.d.	12.97
Kunwee	Domestic	100	200	140	<1	b.d.	12.97
Chiak	Domestic	150	200	150	<1	b.d.	12.97
Pyungc.	Domestic	200	200	150	<1	b.d.	12.98
Pyungc.	Domestic	200	200	150	<1	b.d.	03.99
Kumi		300	2000	1500	<5	b.d.	10.98
Chonju		250	2000	1500	<5	b.d.	12.98
Cheju		1200	3000	2500	<5	b.d.	12.99
Begea	Domestic	3000	150	150	<5	b.d.	12.96
Dulkso	Domestic	850	150	150	<5	b.d.	12.96
Haom Gallery	Domestic	40	120	170	<5	b.d.	09.96
Samsung	Domestic	750	250	150	<5	b.d.	10.96
Inchon	Domestic	50	150	150	<5	b.d.	12.99
Cheju	Domestic	300	-	-	-	-	05.00

7.4.2 The Aduf process by Weir EnVig

The Aduf process was introduced in the late 1980s and early 1990s in South Africa from demonstrations units (0.05–3.0 m^3) to commercial processes (80–2610 m^3) (Table 7.12). The industrial wastewaters treated were soluble or colloidal in nature, with high COD concentrations in the 3.5–37 kg m^{-3} range. The organic removal efficiency was generally > 90%. The system treating the malting effluent was least efficient owing to the presence of non-degradable components such as fatty acids, polyphenols and lipids. As with its aerobic

counterpart, the permeate produced was of a consistently high quality with regard to particulate material, including bacteria and viruses.

Table 7.12: Performance of Weir EnVig Aduf treating various industrial effluents

	Brewery	Maize Processing	Wine Distillery	Malting	Egg Process
Volume (m^3)	0.05	2610	2.4	3.0	80
Operational (months)	3	36	18	5	8
Influent COD (kg m^{-3})	6.7	4.0–15.0	37.0	3.5	8
Permeate COD (kg m^{-3})	0.18	0.3	2.6	0.8	0.35
COD removal efficiency (%)	97	97	93	77	95
Volumetric loading rate (kgCOD m^{-3} d^{-1})	17	3	12	5	6
Specific loading rate (kgCOD kgVSS^{-1} d^{-1})	0.70	0.24	0.58	0.50	0.33
Hydraulic retention time (d)	0.8	5.2	3.3	0.8	1.3
Temperature ($^\circ$C)	35	35	35	35	30
Reactor MLSS(kg m^{-3})	30–50	10	50	10	10–30
Membrane area (m^2)	0.44	9.6	1.75	9.6	200
Membrane Flux (l m^{-2} h^{-1})	10–40	20–40	40–80	20–40	15–30
Inlet Pressure (kPa)	340	500	400	500	500
Liquid velocity (m s^{-1})	1.5	1.8	2.0	1.8	1.8
Tube diameter (mm)	9.0	12.7	12.7	9.0	12.7

Mixed liquor suspended solids (MLSS) concentrations were 10,000–50,000 mg l^{-1}. Ross *et al.*, (1994) argued that sludge concentration was governed by its influence on membrane flux which was fundamentally affected by rheological factors and concentration polarisation. Macromolecules and biomass can form a gel layer on the membrane surface that hinders permeate flow. A lowering of permeate flux can result from high MLSS concentrations as both velocity gradient at the membrane surface and the degree of turbulence of can be decreased by an increase in viscosity. This is the opposite to the situation in aerobic MBRs. Membrane flux was also found to be influenced by the usual operational conditions such as inlet pressure, temperature, linear flow velocity, biological activity and feed biodegradability. Moreover, membrane flux was identified as a reliable indicator for forecasting incipient reactor performance if hydrodynamic conditions remained constant in the membrane modules. Shock influent loads resulted in low fluxes due to a reduction in the microbial membrane cleaning mechanisms.

After 18 months of operation a constant permeate flux of 60 l m^{-2} h^{-1} was maintained in a pilot-plant Aduf treating wine distillery effluent. This flux was sustained for several months before membrane cleaning was necessary. It was hypothesised that an inorganic struvite complex (ammonia magnesium phosphate) had fouled the membrane; a dilute sulphuric acid wash completely restored flux. No irreversible fouling of the membrane has been observed in operational Aduf processes.

7.4.3 The Biorek process by Bioscan A/S

Bioscan A/S has developed the Biorek process for the treatment of agricultural slurry composed of an Aduf, an ammonia stripper and a reverse osmosis (RO) unit, which is capable of purifying the product from the stripper unit to potable water quality. The Aduf process can achieve a greater than 90% COD removal efficiency at a HRT of only 6 d with pig manure as substrate. The sterile product water from the Aduf process is rich in dissolved nutrients. An ammonia stripper removes 95% of the dissolved ammonia and carbon dioxide as an ammonium hydrogen carbonate salt, which can be used as a fertiliser. As a result of carbonate stripping which prevents scale formation on the RO membranes, more than 80% of the water from the slurry is recovered. The RO membrane is used to remove the remaining dissolved salts, namely potassium and phosphate. The remaining liquid concentrate contains mainly these two elements, which can be used as a liquid fertiliser. After first biologically desulpherising the biogas it can fuel a gas motor generator to produce electricity. A commercial 40 m^3 d^{-1} Biorek unit has been in operation in Denmark. Heat from this power production stage is used to heat the Aduf bioreactor and for space heating.

7.5 WEHRLE WERK AG

The Biomembrat process commercialised by Wehrle Werk AG has been treating landfill leachates in Germany since 1990 (Table 7.13). Several of these processes include biological and activated carbon post-treatment, one also incorporates RO, while another uses nanofiltration post-treatment (Wagner and Robinson, 1999). The nine industrial effluents treated by the Biomembrat process include textile, paper and pulp, rendering, tannery, enamelling and pharmaceutical effluents.

Table 7.13: Wehrle Werk Biomembrat processes in current operation

Wastewater Type	Site	Capacity	Number	Start	Comments
Landfill leachate	Europe, mostly Germany	40–900	29	1990–1999	Most include activated carbon post-treatment.
Industrial	Europe	2–4110	9	1994–1999	
Municipal	Germany	140	1	1999	Includes anaerobic and aerobic wastewater treatment
Groundwater	Germany	10	1	1999	Includes activated carbon
Mixed	Netherlands	155	1	1995	Composting effluent and reverse osmosis concentrates from landfill site

7.5.1 Case study: pilot trial on dairy wastewater

When handling dairy wastewater, the success of the biological treatment is influenced by bulk sludge properties, floating sludge foam and large quantities of excess sludge. Conventional construction of dairy wastewater treatment facilities usually requires large areas of land as a result of having to apply low MLSS and long gravimetric separation residence times.

Tests with a Biomembrat pilot plant, using Stork Protech membranes, at a dairy in southern England clearly demonstrated sludge production when compared to existing conventional sewage treatment plants (Wagner and Robinson, 1999). The conventional process used at the dairy was an oxidation ditch followed by gravity setting.

The Biomembrat achieved significantly higher removal efficiencies, with COD effluent concentrations averaging 20 mg l^{-1}, compared with the oxidation ditch which achieved 100 mg l^{-1}. The plant demonstrated that with a sludge load of 0.05 kgCOD kgTS^{-1} d^{-1} the sludge age could be increased to over 100 d resulting in minimal production of excess sludge.
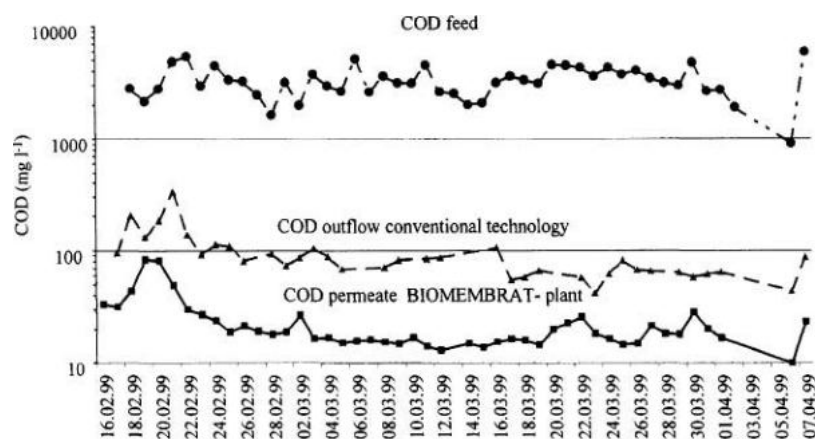
Figure 7.7. Performance of a pilot plant BIOMEMBRAT membrane bioreactor treating a diary wastewater compared to the performance of an oxidation ditch.

7.6 DEGREMONT

As with other commercial MBR processes the ceramic membrane process developed by Degremont was first exploited for the on-site treatment of high strength industrial wastewaters and landfill leachate (Table 7.14).

Table 7.14: Lyonnaise des Eaux Group MBR reference list

Treatment	Location	Capacity (m^3 d^{-1})	Influent
MBR and RO	Arnouville, France	10	Landfill leachate
MBR and RO	Hersin, France	50	Landfill leachate
MBR	Caudry, France	150	Cosmetic industry
MBR and RO	Narbonne, France	10	Landfill leachate
MBR and Ozone	Bagnoles, France	100	Landfill leachate
MBR	Gerenzano, France	50	Landfill leachate
MBR	Rozet, France	800	Dairy industry

The first Degremont MBR was evaluated at a dairy in 1994, when a process of 800 m^3 d^{-1} was commissioned in 1997 (Table 7.15). Though the energy requirement for the MBR process is 60% greater than the conventional activated sludge process the high quality of the effluent was such that 80% of the treated

water was available for reuse. Furthermore, the footprint required for the MBR process was 80% smaller than that for the activated sludge process.

Table 7.15: Comparison of performance of activated sludge process and Degremont MBR treating a dairy wastewater

	Activated Sludge	Degremont MBR
Treated water ($m^3 d^{-1}$)	500	500
Reused water ($m^3 d^{-1}$)	0	400
Bioreactor volume (m^3)	4300	600
Footprint (m^2)	1300	260
COD of treated water (mg l^{-1})	90	50
BOD of treated water (mg l^{-1})	30	<5
SS of treated water (mg l^{-1})	30	0
Energy consumption (kWh m^{-3})	5	8

Like Kubota, Zenon and other MBR manufactures Degremont has identified municipal wastewater treatment as a potential market for their process. As municipal wastewater is relatively dilute compared to industrial effluents the energy consumption for filtration is about 60% of the processes energy requirement. Furthermore, the overall energy requirement can be three to five times greater than conventional activated sludge processes. However, the Degremont MBR can produce a treated water of superior quality compared with the activated sludge process and, along with other commercial MBR processes, widens the spectrum of treated water reuse options (Table 7.16).

Table 7.16: Disinfection capability of the Degremont MBR treating a municipal wastewater

Organism	Number of analyses	Raw water ($100 ml^{-1}$)	Treated water (number of negative results)
Faecal coliforms	26	1.3×10^6	26
Faecal streptococci	26	6.9×10^5	26
Clostridium	26	9.5×10^3	26
Viruses	4	700	4
Giardia cysts	3	7.5×10^3	3
Cryptosporidium oocysts	3	700	3

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8

Costs and conclusions

8.1 COSTS

A major factor preventing the widespread application of MBRs, despite all the process advantages, is capital and operating cost; the most significant of which is the cost of the membrane. Membrane component costs are approximately proportional to plant size as opposed to conventional plants that show an economy of scale. The actual costs of membranes and, more importantly, the fractional importance of membrane replacement on the overall costs are both decreasing. Churchouse and Wildgoose (1999) reported that membrane replacement costs for the Kubota system have decreased from US$400 per m^2 in 1992 to US$100 per m^2 in 2000. More importantly, the relative fraction of the overall cost associated with membrane replacement has decreased from ca. 54% to ca. 9% over the same period. In another study comparing the effects of loading rate and sludge wastage protocol Morimoto *et al.* (1988) reported that a reduction of 20% in membrane investment costs and

[171]

18% in operating costs would be required to make a MBR plant economically competitive for a flow of 100 m^3 d^{-1}.

There are little available data on actual costs reflecting the dynamic nature of the market. However, some cost information has been reported. Mallon *et al.* (1999) described the economics of using a hollow fibre MBR for a design flow of 1800 m^3 d^{-1} for treatment of a chicken processing waste, instead of the currently employed option of pretreatment and discharge to sewer. The capital expenditure of the plant was reported to be UK£1.4 million with operating costs of £0.183 per m^{-3}, representing a pay-back period of approximately 3 years. Judd (1997) reported higher costs at around £1.1–1.4 million per 1000m^3 d^{-1} for a submerged plate and frame system treating a municipal waste. Unlike membrane costs, Davies *et al.* (1998) have shown a downward economy of scale for operating costs such that a decrease in operating costs of 50% was reported as the maximum throughput in a submerged MBR increased from 1.4 to 22.5 Ml d^{-1}. In the case of an anaerobic system, Pillay *et al.* (1994) demonstrated a capital saving of 27% on a 60 Ml d^{-1} plant by incorporating a sidestream membrane to an anaerobic digester rather than using traditional sedimentation.

Davies *et al.*, (1998) compared the economics of a MBR with an activated sludge system for design flows of 2350 and 37,500 population equivalents (p.e.). Total capital costs for the lower flow MBR were reported to be £613,000 of which 78% was attributed to the membrane system (Table 8.1). The reductions in construction and sedimentation offset the costs of the membranes resulting in the activated sludge system being 160% of the capital costs of the MBR plant. However, at the larger scale the difference in the economy of scales between the costs for construction and the membrane units resulted in the conventional systems being only 54% of the MBR plant at approximately £3.9 million.

Table 8.1: Comparison between the costs of membrane bioreactor and conventional activated sludge plant. (Davies *et al*, 1998)

Parameters	MBR (£ sterling)		Activated sludge (£ sterling)	
Treatment capacity (pe)	2350	37500	2350	37500
Max throughput (Ml d^{-1})	1.4	22.5	1.4	22.5
Ave throughput (Ml d^{-1})	0.65	10.5	0.65	10.5
Band screen	59,304	156,085	59,304	156,085
PST/ASP/FST			519,000	1,669562
Membrane system	480,000	6,200,000		
Land	7,000	136,429	21,900	325,573
Civil	126,000	956,095	380,000	1,787,783
Total Capex	613,000	7,292,524	980,204	3,942,259
Manpower	20,000	29,531	20,000	50,000
Chemicals	4,200	5,000	4,200	4,200
Power	10,000	106,917	13,000	148,070
Membrane replacement	30,000	460,653		
Other Overheads	11,173	33,474	19,000	62,460
Annual running costs	75,373	602,101	56,200	264,730
Treatment costs (£ m^{-3})	11.5	6.2	13.2	4.9

PST=primary sedimentation tanks; ASP=activated sludge plant; FST=final sedimentation tanks. Not included in costs: buildings, access road, boundary fences, transfer pumps, pumping stations, storm tanks and storage, sludge disposal and telemetry.

Several studies have attempted to identify the maximum size at which MBRs are competitive with traditional processes (Table 8.2). A very important impact upon the economics of membranes and hence MBRs is the maximum design treatment capacity. Standard practice in the UK is to operate at three dry weather flows (DWFs). This means the MBR operates at a fraction of its capacity for long periods. Davies *et al.* (1998) demonstrated that by reducing the design flow to two DWFs the maximum competitive plant size increased from 12 to 22 Ml d^{-1} even when taking into account the extra storm water storage capacity required. Similarly, a reduction in membrane costs of 50% resulted in a maximum treatment flow of 25 Ml d^{-1} at three DWF. Ogoshi and Suzuki (1999) showed an economic advantage for temporary installations in using MBRs compared to aerobic biological filters at treatment flows < 1000 m^3 d^{-1}. There is no doubt that the maximum competitive size of MBR plants is increasing and will continue to do so. Churchouse and Wildgoose (1999) recently reported that submerged plate and frame systems are now being considered for treatment capacities greater than 50,000 m^3 d^{-1}.

Table 8.2: Summary of maximum economic plant sizes for MBRs

	Country	Compared to	Maximum economical size	Reference
Municipal	UK	Activated sludge	12 Ml d^{-1} @ 3 DWFs 22 Ml d^{-1} @ 2 DWFs	Davies *et al.*, 1998
Municipal	South Africa	Anaerobic digester	7500 kg d^{-1}	Pillay *et al.*, 1994
Temporary installation	Japan	Aerobic biological filter	1000 m^3 d^{-1}	Ogoshi and Suzuki, 1999
Municipal	UK	Activated sludge	50,000 m^3 d^{-1}	Churchouse and Wildgoose, 1999

8.2 CONCLUSIONS

It is arguable that biomass separation MBRs are already established in Japan, America and some continental European countries. In such countries the process has been successfully used for a wide range of applications, but usually for a niche application such as in-building water reuse or leachate treatment. Unique features of the membrane separation bioreactor compared to other biological processes are:

- High-quality effluent, free from solids;
- The ability to disinfect without the need for chemicals;
- Complete independent control of HRT and SRT without the need to select for flocculent micro-organisms or biofilms.

These MBRs also have other key features :

- Reduced sludge production compared to other aerobic processes;
- Process intensification through high biomass concentrations;
- Treatment of recalcitrant organic fractions and improved stability of processes such as nitrification;
- Ability to treat high strength wastes.

The treated water from such MBRs more than meets many of the current quality standards around the world and will meet the increasingly stringent standards of tomorrow. These factors, coupled with continued process optimisation and reduction in membrane costs should guarantees the future of this MBR process. However, research will continue in order to optimise the synergistic relationship between the biological processes and the membrane processes. For example, although low yield has been demonstrated at laboratory and pilot-scale, full-scale systems have yet to take full advantage of this feature.

Both membrane aeration bioreactor (MABR) and EMBR (EMB) technologies have successfully demonstrated the performance enhancement capabilities of incorporating membrane processes in an unusual way into biological wastewater treatment processes.

Membrane aeration bioreactor investigations, at laboratory and small pilot scale, have demonstrated that the process is capable of treating high oxygen demanding wastewaters. Key features of the MABR process that make it an attractive alternative to conventional biological wastewater treatment processes include:

- bubble-less oxygen mass transfer;
- direct transfer of oxygen to biofilm;
- high oxygen utilisation rates;
- high oxygen utilisation efficiencies;

Further demonstration of the technology at large pilot-scale is needed before the process is fully proven. However, a step-change in the efficiency of biological processes could be achieved with the MABR.

In a single-unit operation the EMBR can separate and biodegrade a wide range of chlorinated aromatics from industrial wastewaters. Key features of the EMBR process that make it an attractive alternative to conventional biological wastewater treatment processes include:

- selective membrane extraction of priority pollutants from inhibitory/toxic wastewaters;
- application of specialised microbial cultures;;
- optimisation of the nutrient biomedium to maximise biodegradation without the contamination of undesirable components within the wastewater;
- treated wastewater is not microbially contaminated.

With further development, both MABR and EMB technologies have the potential to enter the portfolio of technologies available for wastewater treatment, particularly the on-site treatment of industrial wastewaters.

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