DEVELOPMENT OF HYBRID ANTIBACTERIAL MEMBRANE BY INCORPORATING SILVER PARTICLE WITH 2,4,6-TRIAMINOPYRIMIDINE AS COMPATIBILIZER

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ABSTRACT

The objective of this study was to develop and characterize the polyethersulfone (PES) incorporated with silver (Ag) as an antibacterial membrane which can remove and disinfect bacteria in a single step for environmental application. The PES-Ag membrane was developed from PES, silver nitrate as an antibacterial agent and 2,4,6-triaminopyrimidine (TAP) as compatibilizer. The influence of $AgNO_3$ loading, molecular weights (MW) of polyvinylpyrrolidone (PVP) as dispersant and type of compatibilizer have been investigated. The resulting membranes were characterized based on their thermal, tensile and structural properties which were used in correlation with the membrane antibacterial properties. The incorporation of Ag in PES membrane has increased the tensile strength doubled as compared to the unmodified PES. Furthermore, it was observed that the highest $AgNO_3$ loading (2 wt%) and the highest MW (360,000) of PVP as dispersant has led to higher silver content on membrane surfaces. This is evidenced from energy dispersive X-ray (EDX) analysis and X-ray photoelectron spectroscopy (XPS). These properties have induced a better antibacterial activity in a disc-diffusion test against *Escherichia coli* (*E.coli*) and *Staphylococcus aureus* (*S.aureus*). The structural characterization by field emission scanning electron microscope (FESEM) revealed that by incorporating TAP as compatibilizer, smaller Ag particles size with improved distribution and average pore size of 0.174 µm was obtained. In addition, the silver residue during fabrication monitored by inductive coupled plasma-mass spectrometry (ICP-MS) was significantly reduced (62.6%). These parameters have led to *E.coli* removal of log reduction value (LRV) 3.59 and 100% growth inhibition tested on *E.coli* suspension of 1×10^6 colony forming unit (CFU/mL). From the adhesion test, this membrane exhibited the least *E.coli* adherence which in turn evidenced its anti-adhesion property. In conclusion, the PES-Ag membrane with TAP as compatibilizer produced was potential in bacteria removal and disinfection below the CFU maximum range for water and waste water treatment.

ABSTRAK

Objektif kajian ini ialah untuk membangun dan mencirikan selaput (membran) poliethersulfona (PES) yang digabungkan dengan perak (argentum) sebagai selaput anti-bakteria yang akan dapat menyingkir dan menyahjangkit bakteria dalam satu langkah untuk aplikasi alam sekitar. Membran PES-Ag dibangunkan daripada PES, garam nitrat perak ($AgNO_3$) sebagai agen anti-bakteria dan 2,4,6-triaminopyrimidine (TAP) sebagai bahan bantu serasi. Pengaruh muatan $AgNO_3$, berat molekul polivinilpirolidone (PVP) yang bertindak sebagai bahan bantu serak (dispersant) dan jenis bahan bantu serasi (compatibilizer) juga telah dikaji. Membran yang terhasil dicirikan bagi menilai sifat terma, kekuatan tegangan (tensil) dan struktur yang kemudiannya dikorelasi kepada sifat anti-bakteria membran tersebut. Penggabungan Ag ke dalam membran PES telah meningkatkan kekuatan tegangan membran dua kali ganda berbanding membran PES tidak terubahsuai. Muatan $AgNO_3$ tertinggi (2 wt%) dan PVP pada berat molekul tertinggi (360,000 Da) didapati telah berjaya menghasilkan kandungan Ag yang lebih tinggi. Ciri ini telah dibuktikan melalui analisis yang menggunakan kaedah spektroskopi penyebar tenaga sinar-X (EDX) dan kaedah spektroskopi elektron-foto sinar-X (XPS). Sifat-sifat ini seterusnya telah mencetuskan sifat anti-bakteria yang lebih baik, dibuktikan melalui ujian pembauran-cakera (disc-diffusion) terhadap bakteria *Escherichia coli* (*E.coli*) dan *Staphylococcus aureus* (*S.aureus*). Pencirian struktur dengan menggunakan mikroskopi imbasan elektron pemancaran medan (FESEM) telah memberi maklumat bahawa dengan menggunakan TAP sebagai bahan bantu serasi, partikel Ag yang lebih kecil dengan taburan yang lebih baik pada saiz liang purata 0.174 μm telah dperolehi. Di samping itu, sisa Ag yang terlarut resap (leach) semasa pembuatan membran yang dikawal dengan menggunakan spektrometri jisim-berganding plasma teraruh (ICP-MS) didapati menurun dengan nyata sebanyak 62.6%. Keseluruhan parameter yang dikaji telah menunjukkan bahawa penyingkiran *E.coli* adalah pada nilai penurunan log (LRV) 3.59 dan 100% perencatan pertumbuhan apabila diuji pada 1×10^6 unit koloni terbentuk per mL (CFU/mL). Membran ini juga didapati menunjukkan lekatan bakteria (bacterial adherence) yang terkecil dalam ujian lekatan terhadap *E.coli* sekaligus membuktikan sifat anti-lekatan. Kesimpulan daripada kajian ialah membran PES-Ag dengan TAP sebagai bahan bantu serasi adalah sangat berpotensi dalam penyingkiran dan perencatan bakteria di bawah julat CFU untuk air dan rawatan air.

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LIST OF ABBREVIATIONS

MF	-	Microfiltration
DBPs	-	Disinfection by-products
DOM	-	Dissolved organic matter
UF	-	Ultrafiltration
LRV	-	Log-reduction value
UV	-	Ultra-violet
E.coli	-	*Escherichia coli*
S.aureus	-	*Staphylococcus aureus*
DNA	-	Deoxyribonucleic acid
TGA	-	Thermogravimetric analysis
DSC	-	Differential scanning calorimetry
XRD	-	X-ray diffraction
XPS	-	X-ray photoelectron spectroscopy
EDX	-	Energy dispersive X-ray
ICP-MS	-	Inductive coupled plasma-mass spectrometer
CFU	-	Colony forming unit
NOM	-	Natural organic matter
DOC	-	Dissolved organic carbon
SS	-	Suspended solids
TDS	-	Total dissolved solid
pH	-	$-\log[H^+]$
WHO	-	World of Health Organization
DLVO	-	Deryaguin-Landau and Vervey-Overbeck
MWCO	-	Molecular weight cut-off
Da	-	Dalton
AFM	-	Atomic force microscopy
ATR-FTIR	-	Attenuated total reflection-Fourier transform infra-red spectroscopy
FESEM	-	Field emission scanning electron microscope
ppt	-	Part per trillion
ppm	-	Part per million
PWP	-	Pure water permeation
TOC	-	Total organic carbon
MW	-	Molecular weight
NA	-	Nutrient agar
OD	-	Optical density
MD	-	Minimal Davis
rpm	-	Revolution per minute
v/v	-	Volume per volume
Fig.	-	Figure
MTS	-	Mechanical testing system

Wt.	-	Weight
Eq.	-	Equation
SD	-	Standard deviation
DMFC	-	Direct methanol fuel cell
cps	-	Count per second
BE	-	Binding energy

LIST OF SYMBOLS

%	-	Percent
˚C	-	Degree Celcius
h	-	hour
mA	-	miliampere
kV	-	Kilo Volt
°	-	degree
θ	-	theta
µM	-	Micro molar
wt. %	-	Weight percent
d	-	Diameter
J_v	-	Pure water permeation (Flux)
r_m	-	Mean pore diameter (µm)
ε	-	Porosity (%)
Q	-	Volume of permeate per unit time (m^3s^{-1})
A	-	Membrane surface area (m^2)
ΔT	-	Permeation time (s)
µm	-	Micrometer
nm	-	Nanometer
C_p	-	Concentration of permeate (ppm)
C_f	-	Concentration of feed (ppm)
mg/L	-	Miligrams per litre
mm/min	-	Milimeter per minute
V_R	-	Repulsive energy
V_A	-	Attractive energy
V_T	-	Total energy
T_g	-	Glass transition temperature

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CHAPTER 1

INTRODUCTION

1.1 Research Background

Microbiological contamination of water sources has long been a concern to the public. According to some authors, there were numbers of various bacterial species available (ranging from 10^2 to 10^4 mL^{-1}) in raw water as well as sewage effluents (Bonnélye *et al*., 2008; Goldman *et al*., 2009) tend to adhere to surfaces and grow mainly at the expense of nutrients accumulated from the water phase. Microbiological contamination in any sources should be avoided at any cost since in the production of potable water, only a limited number of bacteria (depends on the type of bacteria) are acceptable. The process for the removal of contaminants depends not only on the nature of the microorganisms but also on the desired levels of purity.

The use of membrane filtration in water treatment has greatly contributed to greener technology. For example, microfiltration (MF) membrane has been widely applied in water purification process due to its capability to remove microorganisms and to treat harmful pollutants as well as dissolved organic matters (DOM) (Ghayeni *et al*.,1996; Oh *et al*., 2007). Laine *et al.* reported that ultrafiltration (UF) applications represent 74% of the total installed low-pressure membrane full-scale plants (identified world-wide) in water industry in order to meet more stringent regulations in producing drinking water (Laine *et al*., 2000).

It was reported in the open literature that membrane technology is one of the disinfection technique where microorganisms are retained without any chemicals engagement. However, the problem of biofouling aroused when membrane is applied, due to the accumulation of microorganisms on membrane surfaces. In addition, the current practice of membrane filtration required additional step addressed as disinfection step via techniques such as chlorination (the most common one), ozonation and UV. There were many and thorough discussion available in the use of chlorination recently. The use of chlorination may lead to the disinfection by-products (DBPs) release which in turn exposed consumers to potential carcinogenic compounds such as the derivatives of chloramines.

Many studies have been conducted to overcome/meet the restrictions as well as to resolve membrane fouling problems due to the uncontrolled accumulations of micro-species. In handling biofouling problems, membrane modification, low-flux operation and chemical cleaning are areas to be explored (Chang *et al*., 2002; Khor *et al*., 2007). The effective prevention of microbial growth in a membrane system can only be achieved when continuous and sufficiently high chlorine concentration is maintained. However, due to stricter legislative regulation on chlorine usage, other effective and environmental-friendly alternative is needed.

In membrane modification, the research in combining inorganics into polymer matrices has been expanding since 1990-s. The inorganics chosen were tailored with the application such as catalysis, biochemistry, separation and sensing. In gas separation, the inorganic fillers namely zeolite, carbon molecular sieve, silica and metal oxides has contributed to enhance membrane separation performance specifically in addressing flux decline and selectivity (Rafizah *et al*., 2008; Kusworo *et al.*, 2008; Ismail *et al*., 2009; Mataram *et al*., 2010).The combination of inorganics in polymer matrices or well-known as mixed-matrix provides the solution for highly cost-maintenance and brittleness of inorganic membranes. The inorganic fillers in mixed matrix membrane act to create preferential permeation pathways for selective permeability while posing a barrier for undesired permeation in order to improve the separation performance (Goh et al., 2011).

In water application, attempts were made in addressing flux decline due to the accumulation of (micro- or macro-)species onto membrane surfaces which in turn affecting the separation performance. The important issue in membrane manufacturing is to develop membrane with suitable pore size in order to attain various sizes of contaminants. In addition, membrane must also show sufficient resistance towards the feed components as well as the operating condition. In antibacterial application, a number of researches have been conducted in exploring silver-incorporation to polymeric materials such as cellulose acetate (CA), polacrylonitrile (PAN), polysulfone (PSf) and chitosan for the application of water treatment, nano-fibre and food-packaging (Chou *et. al*., 2005; Wang *et. al*., 2005; Ma *et. al*., 2008, Zodrow *et. al*., 2009). Silver was found to leach in the reported articles and to date; attempts on overcoming this problem are still not published in the open literature.

1.2 Problem statements

The conventional UF membrane in water and waste water treatment established in bacteria removal has achieved a promising rejection value of >99% or to be specific, a log reduction value (LRV) of >3. However, after the membrane filtration process, it is necessary to perform an extra step which is disinfection as a secondary bacteria control barrier and distribution system protection (Ghayeni *et al*., 1996; Zio *et al*., 2005). The options available for disinfection are: UV, ozonation and chlorination. The most commonly used method is chlorination due to the easy-handling process and cost effectiveness. However, the major drawbacks of this method is when greater number of bacteria present, higher concentration of chlorine is needed, hence more disinfection by-products (DBPs) will be released in the water distribution system. Current waste water treatment with microbial burden is facing the problem of biofouling due to the accumulation of microorganisms over operation time. In order to address the issues, current research is conducted to explore the possibility and effectiveness of using a UF membrane incorporated with an antibacterial agent in removing bacteria as well as to investigate its potential in behaving anti-biofouling properties.

1.3 Objectives of the study

Based on the existing problem statements, the current study has been performed with the following objectives:

i. To develop an antibacterial membrane by incorporating silver (Ag) as an antibacterial agent without sacrificing membrane fluxes and removal abilities and to characterize the membrane in terms of mechanical, morphological, water permeation, hydrophilicity and pore sizes.
ii. To study the effect of incorporating a compatibilizer, PVP of different molecular weights in membrane properties and performances.
iii. To evaluate the effect of incorporating different compatibilizers in membrane properties and performances.
iv. To evaluate the fluxes of prepared membranes using pure water permeation test on the custom-made test-rig.
v. To evaluate the antibacterial performance of membrane using disc diffusion method, filtration of bacterial suspension and anti-biofouling tests.

1.4 Research scopes

In order to achieve the above mentioned objectives, the following scopes of study were drawn.

i. Preparation of dope using PES as polymeric material, NMP as solvent and $AgNO_3$ as an additive or precursor of antibacterial agent, silver (Ag).
ii. Selection of $AgNO_3$-loading and compatibilizer based on the evaluation in the miscibility and antibacterial tests.
iii. Fabrication of PES-$AgNO_3$ asymmetric membrane using phase inversion technique and characterization of membranes in terms of mechanical strength, hydrophilicity, overall porosity, pore sizes and water permeation.
iv. Evaluation of Ag-entrapment in prepared membranes by using ICP-MS, EDX and XPS techniques.
v. Membrane fluxes measurement was carried out by using custom-made test rig at pressure range 1-6 bar.

vi. Performance measurement of prepared membranes was conducted in terms of antibacterial activity by using disc diffusion method and bacteria removal via the filtration of bacterial suspension.

vii. Performance measurement of prepared membranes was conducted in terms of anti-biofouling properties through an anti-adhesion test.

viii. Comparison of PES pristine membrane with PES antibacterial membrane in all characterization and antibacterial tests.

1.5 Research significance

This study is of significance to the research of water treatment which involves disinfection steps. The antibacterial membrane extends the multi-steps options for water treatment to a stand-alone removal and disinfection of bacteria. The results obtained in the study also provide the information in bacteria-removal and bacteria-killing mechanisms which lead to the most effective options in treating polluted water. Furthermore, the information on silver entrapment obtained in this study would be beneficial to the other related fields such as in medicinal and electrical field where silver is optimized in wound dressings and conducting material.

1.6 Organization of the thesis

The thesis is divided into six chapters. The first chapter presents the research background as well as the problem statement. The research objectives, scopes and significance are also highlighted in first chapter. Chapter two provides the literature review on bacteria removal which includes the theories of the whole process and the options available for bacteria removal. The advantages of antibacterial membrane, current status and future direction of the technology are also discussed in this chapter. Chapter three is dedicated to the detailed description of the research methodology. The material selection for dope preparation, membrane fabrication and performance testing conducted in this work are explained in this chapter. In chapter four, the effect of silver content on the properties and performance of fabricated membranes are explored and discussed. Subsequently, Chapter five describes the effect of polyvinylpyrrolidone (PVP) of various molecular weights on antibacterial properties of the resulting membranes. Chapter six discusses the significant contribution of different compatibilizers in membrane antibacterial activities. Other improved properties and the comprehensive discussions on the anti-adhesion properties of resultant membranes are also included. Finally in Chapter seven, conclusion of the research is drawn and the potential future works are proposed.

CHAPTER 2

ANTIBACTERIAL MEMBRANE FOR BACTERIA REMOVAL: A REVIEW

2.1 Introduction

In conventional treatment of contaminated water sources, several methods have been applied to provide multiple barriers to the spread of pathogenic microorganisms and have minimize the spread of waterborne disease. Table 2.1 lists each treatment which can either be used individually or couple with other methods in a multi- step process.

Amongst the processes listed, sedimentation is considered as an easy and widely accepted technique due to its natural principle which uses earth gravitational force to settle down particles/suspended solids (SS) from a suspension. The process consumes low energy therefore reduce the processing cost. However, some drawbacks quoted by Guazzelli (1984) was the low loading rates which is only 1- 2 $m^3m^{-2}hr^{-1}$ plus large space needed, hence higher construction cost will be required. Other than the natural force, sedimentation applies coagulants as an aid to cater high sludge loading. The efficient sweep coagulation can only be achieved at higher coagulant dose. Excessive slime bacteria which result in filter clogging are another drawback in sedimentation (Horan and Mara, 2003).

Table 2.1: Available treatment methods in tackling the spread of pathogenic microorganisms

Treatment method	Processes	Effectiveness	References
Coagulation	Coagulation or coagulation-flocculation is a conventional technique to remove organic and inorganic suspension, colloids and other natural organic matter (NOM). This process uses chemicals addressed as coagulants and coagulant aids. Examples of common coagulants are $Al_2(SO_4)_3$ (alum), $Fe_2(SO_4)_3$ (copperas), $FeSO_4$ and $FeCl_3$ while examples of the aids are bentonite, $(Al_2O_3)4SiO_2.2H_2O$, sodium silicate, Na_2SiO_3, lime, $Ca(OH)_2$ and calcium carbonate, $CaCO_3$. Primary coagulants neutralizes the electrical charges of particles in the water which cause the particles to clump together, while the aids are generally used to reduce flocculation time and specifically used for clear water with very low turbidity that does not coagulate well with usual procedures.	Removes • Turbidity at ~ 99% • NOM (in terms of dissolved organic carbon, DOC (up to 59.5%) • Bacteria at >3 log reduction value, LRV	(Konieczny et al., 2009) (www.thewatertreatments.com) (Qin *et al*., 2006)
Sedimentation	Sedimentation is a physical water treatment process to settle suspended solid that has been deposited by natural processes in water under the influence of gravity. The settling rate of sedimentation can be dramatically improved by the addition of small dosages of polymeric flocculants. The higher the solid concentration, the faster the flocculation occurs and hence larger flogs will be produced.	• *E.coli* removal of >50% (depending on turbidity and the characteristics of particular sediment). • For 50 mL of *E.coli* suspension, population was reduced to 1/120, 1/100,000 and 1/1,400,000 after 24, 48 and 72 h respectively.	(Gutbai and Gregory, 1991) (Milne *et al*., 1986) (Kawabata & Tanabe, 2005)

Filtration	Filtration is a process to separate matters from fluid by passing the mixture through a porous media that entraps the solids in its matrix or retains them on its surface. The amount of removal is a function of the filtering media. The removal was subject to several factors including mechanisms (straining or adsorption), the grain size of porous media, organic matter content, bacteria species and etc.	• The median reduction is 10^4. • Soil-filtration has reduced total bacteria up to 99.94%. • Filtration of *E.coli* suspension on an antibacterial membrane has resulted in 100% growth inhibition.	(Sterik *et al*., 2004) (Vanderbroucke *et al*, 1995) (Gilbert *et al*., 1976) (Basri *et al*., 2010)
Disinfection	Disinfection is considered as a primary mechanism for the inactivation of pathogenic organisms to prevent the spread of waterborne disease to downstream users and the environment. Disinfection is performed via physical or chemical techniques. Radiation, filtration and heating are some examples of physical disinfection while chlorination and ozonation are chemical disinfecting method.	By UV disinfection, the $\log_{10}\frac{N_0}{N}$: • *E.coli* 3.8 • Total coliform 3.3 • Enterococci 3.3	(Madaeni, 1999) (Olanczuk-Neyman *et al*., 2001)

Sedimentation and further flocculation were needed to produce good quality water while the use of powder activated carbon (PAC) was possibly needed to remove taste and odor (Hagen, 1998). In order to complement the conventional water treatment processes, disinfection was performed by using chemical disinfectants or physical methods. The objective of disinfection is to render an object or field free from infection in which the infection may represent a risk to persons or environment (Gilbert and Brown, 1995).

2.2 Disinfection in water and wastewater treatment

Diseases caused by pathogenic bacteria, viruses, protozoa or helminthes are the most common and wide-spread health risk associated with drinking water. For this reason, the World of Health Organization (WHO) has placed the greatest importance on the microbiological quality of drinking water emphasized the potential consequences of microbial contamination are such that its control must never be compromised (Gorchev, 1996). To the least, water sources must be protected from contamination by human and animal wastes which contain a variety of bacterial, viral, protozoa and helminthes pathogens which are the sources of the waterborne disease. The characteristics of the main waterborne disease source are listed in Table 2.3.

Waterborne disease is disease resulted from improper sanitary disposal of human feces. The feces of healthy persons contain 1 to 1000 million per gram of each of the following groups of bacteria: enterobacteria (e.g *E.coli*), enterococci, lactobacilli, clostridia, bacteriodas, bifidobacteria and eubacteria (Hammer and Hammer Jr, 2008).

In order to meet the stringent regulations by WHO, disinfection methods (chemical and/or physical) has been adopted in water and wastewater treatment. The commonly used disinfectants are listed in Table 2.2 with their risks, advantages and efficiency against different contaminants for general comparison. Several studies have shown that the efficiency of UV as a disinfection method is highly dependent on the concentration of SS (Narkis *et al.*, 1995; Hurst, 1996; Blume *et al.*, 2002).

This is due to the fact that SS can protect the bacteria through a 'sheltering' flogs which prevented the UV-light from penetrating and destroyed by the disinfectants (LeChevallier, 1988). The UV-light cannot penetrate large particles from ~50 μm in diameter, thus the required energy will be raised drastically (Neis and Blume, 2003).

Table 2.2: The difference between bacteria, viruses and protozoa (Madigan *et al*., 2000).

Organism	Size	Description	Examples (waterborne)
Viruses	20-120 nm	Biological agents consisting of molecules of nucleic acids and protein envelope.	Enterovirus, coxsackievirus, echovirus, rotavirus, hepatitis A & B.
Bacteria	1-6 μm	Unicellular and organism with nucleus	*E.coli*, *Salmonella sp*., *Shigella sp*.
Protozoa	*Cryptosporidium sp.* (4-6 μm) *Giardia sp.* (8-12 length) × (7-10 μm width)	Protozoa are single-celled eukaryotes (organisms whose cells have nuclei) that show some characteristics usually associated with animals, most notably mobility and heterotrophy	*Giardia duodenalis*, *Cryptosporidium sp*., Entamoeba

Table 2.3: Comparison of commonly used disinfectants in water reclamation (Asano *et al*., 2007)

Characteristics	Chlorine gas	Sodium hypochlorite	Chlorine dioxide	ozone	UV radiation
Deodorizing ability	High	Moderate	High	High	na*
Interaction with organic matters	Oxidizes organic matter	Oxidizes organic matter	Oxidizes organic matter	Oxidizes organic matter	Absorbance of UV irradiation
Corrosiveness	Highly corrosive	Corrosive	Highly corrosive	Highly corrosive	na*
Toxic to higher forms of life	Highly toxic	Highly toxic	Toxic	Toxic	Toxic
Penetration of particles	High	High	High	High	Moderate
Safety concern	High	Moderate to Low	High	Moderate	Low
Solubility	Moderate	High	High	High	na*
Stability	Stable	Slightly unstable	Unstable	Unstable	na*
<u>Effectiveness as disinfectant</u>					
Bacteria	Excellent	Excellent	Excellent	Excellent	Good
Protozoa	Fair to poor	Fair to poor	Good	Good	Excellent
Viruses	Excellent	Excellent	Excellent	Excellent	Good
Byproduct formation	THMs and HAAs	THMs and HAAs	Chlorite and chlorate	Bromate	None known in measurable concentrations
Increases TDS	Yes	Yes	Yes	No	No
Use as a disinfectant	Common	Common	Occasional	Occasional	Increasing rapidly

*na = not applicable

The recent development in the area of disinfection has been discovered to reflect a great diversity and complexity of product. As an example, for chlorination, chlorine in its free form may react with a group of organic acid available in water and result in trihalomethanes (THM) or other DBPs formation (Asano *et al*., 2007).

Basically when chlorine in gas form is added to water, hydrolysis molecules will occur and hypochlorous acid (HOCl) will be formed (Smethurst, 1988):

$$Cl_2 + H_2O \rightarrow HOCl + H^+ + Cl^- \quad (2.1)$$

Because hypochlorous acid is a very active oxidizing agent, it may also react with nitrogen derivatives for an example ammonia and form chloramines;

$$NH_3 + HOCl \rightarrow NH_2Cl \text{ (monochloramine)} + H_2O \quad (2.2)$$

$$NH_2Cl + HOCl \rightarrow NHCl_2 \text{ (dichloramine)} + H_2O \quad (2.3)$$

$$NHCl_2 + HOCl \rightarrow NCl_3 \text{ (trichloramine)} + H_2O \quad (2.4)$$

The equations (2.1-2.4) are the pathways that show the DBPs formation resulted from chlorination. The action of bacteria-killing by chlorine was due to the direct toxic action not only by chlorine in its free form but also by hypochlorous acids and chloramines (Smethurst, 1988). Other than the DBPs formation, Scholz (2006) also highlighted the disadvantage of chlorination which is the ‘chlorine taste’ of water and the need of extra care of storage and handling.

Effective prevention of microbial growth in water treatment systems can only be achieved when a continuous and sufficiently high chlorine concentration is maintained. However, this condition cannot be viewed as an ultimate solution, considering growing environmental concerns and stricter legislative regulations regarding the discharge of chlorinated brines. Therefore, membrane materials with reduced bacterial affinity have been actively researched (Flemming, 1997). The investigation by Hagen (1998) revealed that the present disinfection and filtration methods should be replaced by a more suitable membrane filtration process.

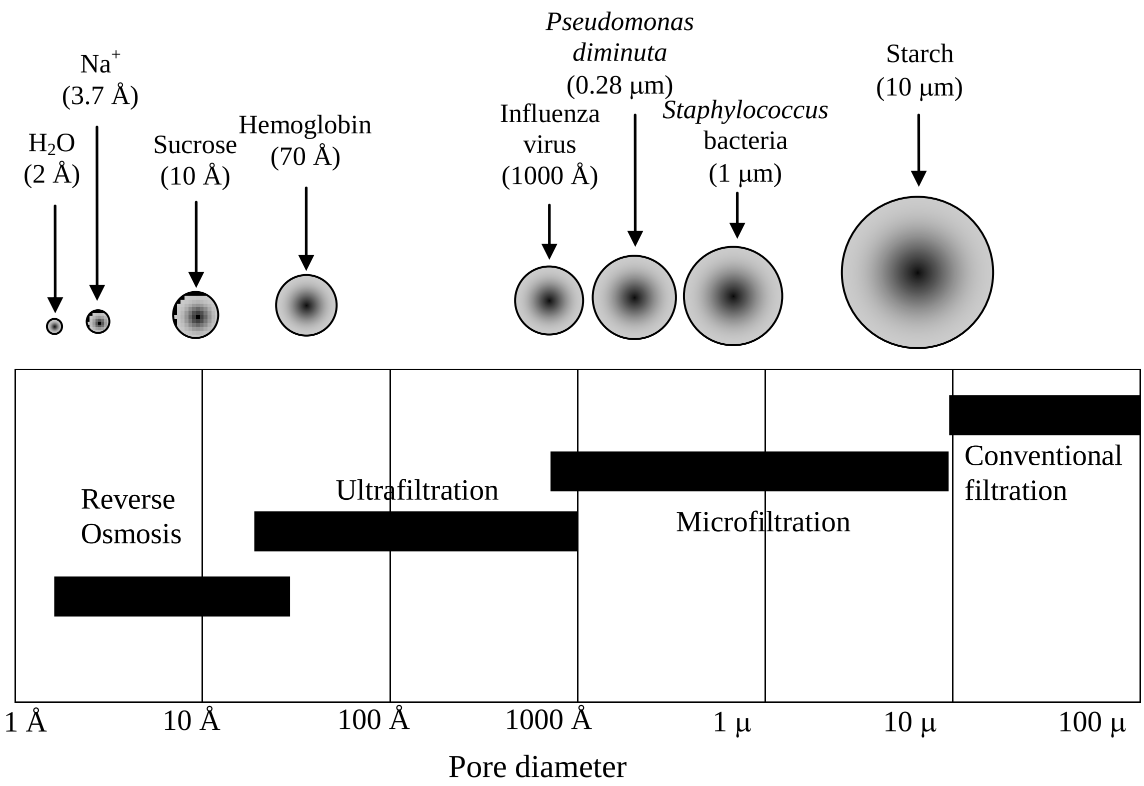

Figure 2.1: Pressure driven membrane processes classified principally by average pore diameter (Eykamp, 1995).

2.3 Membrane technology in bacteria removal

In general, membrane is a barrier that separates two phases and restricts the transport of various species in a specific manner when a driving force is applied. In other words, when driving force is applied, the membrane placed in a fluid system will retain one component by sieving or size-exclusion mechanism and produce purified solution. Pressure-driven membrane processes which are reverse osmosis, nano-, ultra- and microfiltration are now being extensively used for the purification of natural and waste waters. Figure 2.1 illustrates the pore size range of pressure driven membrane processes that are used to separate particles of various size range.

Ultrafiltration (UF) is a pressure driven membrane process whose nature lies between nanofiltration and microfiltration (MF). MF is typically known and used for turbidity reduction and removal of suspended solids within the approximately size of 1-30 µm (Li *et al*., 2003) meanwhile UF membranes are commonly used to remove some viruses, color, odor, and some colloidal natural organic matter. Both processes require low transmembrane pressure (1- 30 psi) to operate, and both are now being used as pretreatment in desalination processes such as reverse osmosis, nanofiltration, and electrodialysis. As a pressure driven membrane process, UF membrane normally possesses asymmetric structure with thin but relatively dense top layer (thickness 0.1-1.0 µm), supported by a porous substructure (thickness ≈50-150 µm) in which suspended colloids and particles in the approximate size range of 10-1000 Å are retained. An illustration in Figure 2.2 simplified the separation concept in membrane. Although UF has been commonly used in current market, the improvement on the available system is still necessary.

Transportation of molecules or particles via membrane occurs due to the driving forces applied. This driving force can be chemical potential gradient, e.g concentration gradient or pH gradient; pressure difference; electrical potential difference or combination of these (Mulder, 1991). In bacteria removal, bacteria transport is generalized to occur by advection, diffusion (for small bacteria) and chemotaxis (Corapcioglu, 1996). Chemotaxis is the preferential movement of bacteria in response to chemical gradients such as areas of higher nutrient concentrations.

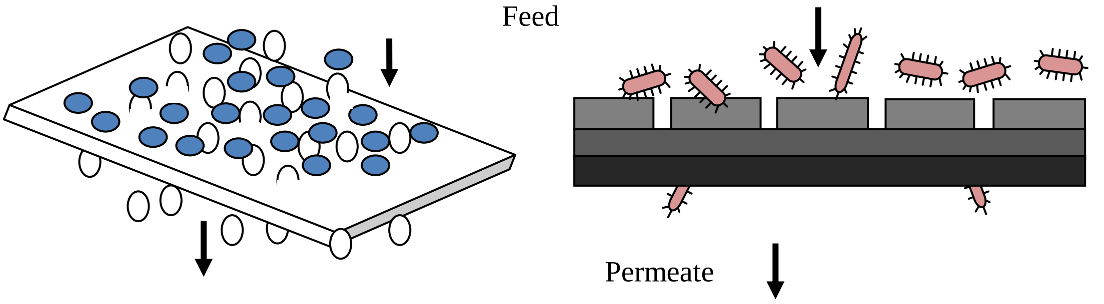

Figure 2.2: Simplified concept schematic of membrane separation. A desired component (water) is allowed to pass through while non-desired component (bacteria) is retained.

When membrane with smaller pore size is used or bigger solute species need to be retained, the higher pressure has to be applied in the operating system. The proportional relationship between the membrane fluxes and the driving force is:

$$\text{Flux} = \text{Proportionality factor} \times \text{Driving force}$$

$$J = A \times X \tag{2.5}$$

where *A* is the proportionality factor determines how fast the components or particles is transported through the membrane. In other words, *A* is a resistance measurement exerted by membrane as a diffusion medium when force is applied to the components or particles.

According to Song and Elimetech (1995), the net velocity of bacterial cell normal to the membrane surface is largely determined by normal convection with small contributions by tangential correction and Brownian diffusion. The interaction force profile suggested that aggregation were enhanced in acidic medium even though the bacterial deposition rate is lower. The model studied also suggested that the increment of permeation velocity resulted in higher bacterial deposition rate.

2.4 Physicochemical interaction between bacteria and surface

The physicochemical interaction between bacteria and surfaces has been highlighted. The schematic in Figure 2.3 illustrates the interaction among bacterial cell, inorganic particles and the surface of porous media. Removal of bacteria from the flowing liquid phase generally occurs by filtration and adsorption or cell death (Corapcioglu, 1996). The various bacterial attachment and detachment mechanisms are affected by one or more factors as listed in Table 2.4. According to Yuehuei and Friedman (2000), bacteria surface hydrophobicity is an important physical factor for adhesion. Generally, hydrophobic bacteria prefer materials with hydrophobic surfaces while hydrophilic characteristics prefer hydrophilic surfaces. However, hydrophobic bacteria adhere to a greater extent than hydrophilic bacteria.

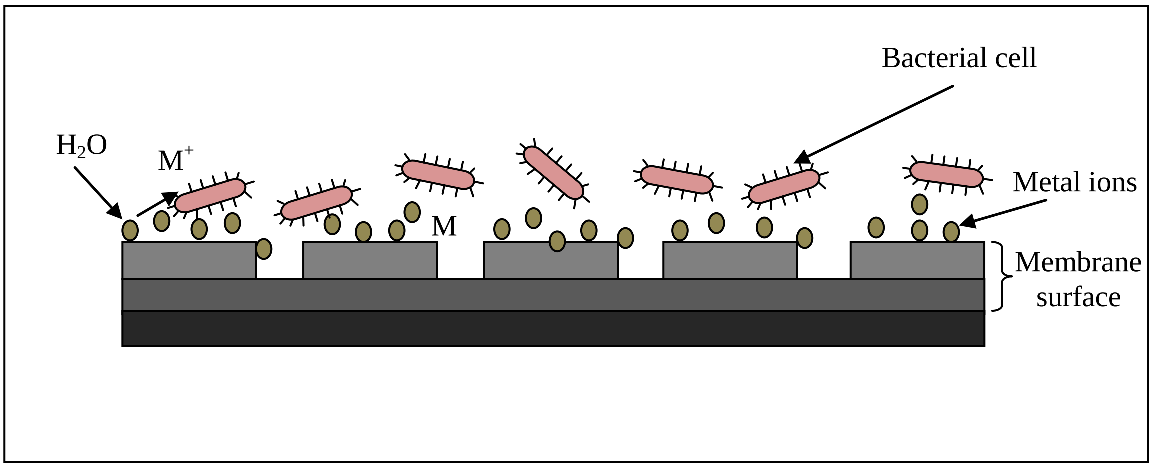

Figure 2.3: Schematic of the physicochemistry between bacteria-nanoparticles and membrane matrices.
(Yuehei and Friedman, 2000)

When approaching the entrance of a pore, Leblue *et al*. (2009) explained that bacteria are submitted to the shear and drag forces created by the trans-membrane pressure (TMP) applied during the filtration step. Such stress may lead to bacteria volume reduction and surface deformation (governed by the cell wall Young Modulus value) which would allow the cell to penetrate into the membrane pore. Whether the cell membrane is disrupted or not, the bacteria still have the possibility to pass the pore. If the bacteria penetrate the membrane and retain its integrity, these bacteria will keep their pathogenicity and hence risk consumers. To address the problem, the inorganic antimicrobial agent attached on the membrane surfaces and in membrane pores will act and perform biocidal action. The system which combined membrane technology with inorganic antimicrobial is efficient in such a way that the metal ion is bound within a delivery system that stabilizes them and then releases them through a process of ion exchange at the surface (Peinemann and Nunes, 2010).

Table 2.4: Factors affecting bacterial attachment and transport (Corapcioglu, 1996)

Factor	Effect on transport or attachment
pH	An attachment favors low pHs.
Ionic strength	Attachment increases with higher ionic strength due to the 'particle double-layer' size reduction.
Clay-content	Attachment increases with high clay content due to larger specific area for adsorption.
Oxygen limitations	Oxygen-limited biofilms exhibit lower shear removal rates but higher sloughing.
Change on media	Attachment of negative bacteria will be high in positive charges media.
Flow rate	Higher flow rates reduce bacterial attachment.
Nutrient concentration	Bacterial size reduced in higher nutrient concentrations.
Bacterial size	Smaller bacteria may interact with media less and may not be removed by filtration as easy as bigger bacteria. On the other hand, larger bacteria have been shown to move faster than small bacteria.
Cell concentration	At low cell density, attachment is favored. Bacteria tend to move from high concentration areas to low concentration areas by a tumbling diffusive flux.
Bacterial motility	Motile bacteria may migrate faster than non-motile bacteria through chemotaxis.
Water content	Bacteria moves faster through unsaturated soil at higher water content.

2.5 Bacteria retention in porous media

According to Dunne Jr., (2002) the process of bacterial adhesion is dictated by the variables including the species of bacteria, surface composition, environmental factors and the essential gene products. From an evolutionary standpoint, the selective advantage of bacterial adhesion was postulated to favor a nutritional and non-hostile environment and provide a level of protection. Frimmel *et al.* (2007) discusses the two types of deposition mechanism namely straining and interception. Straining is generally about retaining big agglomerates while interception is about retaining small aggregates on porous surface after collision. The interception mechanism which is dominated by physicochemical interactions between the cell surface and the porous medium has been reported to govern the adhesion of cells. In order to understand the process of bacterial adhesion, the two mechanisms in bacteria removal are discussed.

2.5.1 Bacteria retention via straining

According to Stevik *et al*. (2004) straining mechanism depends on the grain size of the porous media. Generally, the extent to which the bacteria are retained by straining is inversely proportional to the size of the filter media particles. In other words, the smaller the filter media size, the more bacteria will be retained via straining. By considering filter-media factors, straining will become a dominant mechanism when the average cell size of the bacteria is greater than the size of 5% of the grains that compose the porous material (Stevik *et al*., 2004). The presence of macropores in filtration has been found to result in poor volume utilization and allow a more rapid and distant bacteria movement (Chandler *et al*., 1981). In brief, the transport for most of the bacteria in a system on saturated flow (e.g waste water treatment) is found to favorably take place in the smaller pores. Generally, larger cells will be more efficiently removed by filtration.

Weiss *et al*. (1995) studied the effect of bacterial cell shape on the transport in porous media and suggested a preferential removal of long, rod-shaped cells during transport. Bacterial straining can also be influenced by flow rates and hydraulic loading. A high flow-rate may increase the average water suction in an unsaturated filter medium. As a result, greater transport may occur via larger pores which in turn decrease the effect of bacterial straining by porous material. Ausland *et al*. (2002) observed a higher removal of fecal coliform bacteria in filtration systems using uniform pressure distribution as compared to gravity dosing. Another factor to be considered in bacteria removal via straining is clogging (Vandevivere and Baveye, 1992). Clogging occurs due to the biomass growth in the porous media. Bacteria removal is more efficient in clogged filtration system due to the hydraulic disfunction which diminishes the purification of wastewater (Bouwer, 1974; Gannon *et al*., 1991).

2.5.2 Bacteria retention via adsorption

In contrast to straining mechanism, adsorption is the dominant mechanism in bacteria retention when media pores sizes are larger than that of bacteria (Sharma *et al*., 1985). The bacterial adsorption on solid surfaces involves two stages mechanisms that conform to the classical Derjaguin-Landau and Verwey-Overbeek (DLVO) theory which has been first suggested on charged colloidal particles. The stages were illustrated in Figure 2.4. The first stage is a reversible mechanism controlled by electrostatic interactions between the cell surface and the adsorbent (porous media). Weak interaction is present between the bacterium and porous material. During this stage, reversibly attached bacteria can detach from the surface of a particle and return to the water phase, depending on the conductance and chemical properties of the fluid or aqueous solution.

In second stage, bacterial adsorption forms a much more persistent bond between adsorbent (porous media) and adsorbate (bacterium). This mechanism is irreversible and sometimes referred as adhesion (Olson *et al*., 1991). According to the classical DLVO theory, the energy of interaction (V_T) between a bacterium and solid surface is (Derjaguin and Landau, 1941):

$$V_T = V_R + V_A \tag{2.6}$$

V_R = repulsive energy resulting from the overlapping of the electrical double layer of cell and substratum (generally repulsive), V_A = attractive energy resulting from van der Waals interactions (generally attractive).

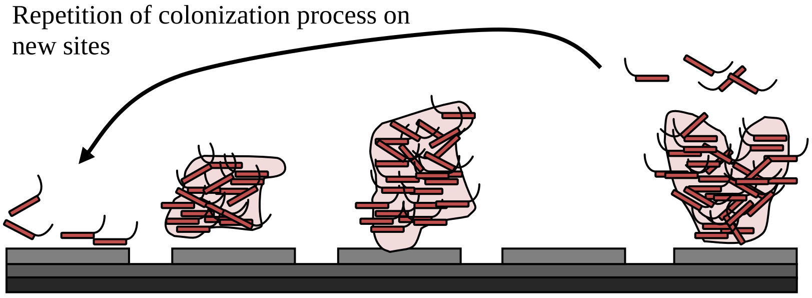

Figure 2.4: Stages of biofilm formation and the two-stages bacterial adsorption (Katsikogianni and Missirlis, 2004; Houdt and Michiels, 2005).

According to Hermansson, who extended the classical DLVO theory, V_A (Hermansson, 1999):

$$V_A = -\frac{Ar}{6d} \tag{2.7}$$

where A is the Hamaker constant, d is the separation distance between the cell and the solid surface and r is the cell radius (assuming cells are spherical). The DLVO theory states that the distance of separation between colloidal adsorbents (porous media) and adsorbates (bacterium) is the distance at which the repulsive (V_R) and attractive (V_A) energy are balanced.

Hori and Matsumoto (2010) explained the two steps mechanism in DLVO theory by relating them to the ionic strength as illustrated in Figure 2.3. At low ionic strength, the energy barrier prevents the bacterium from approaching solid surfaces via Brownian motion.When the energy barrier becomes higher and further from the solid surface (at lower ionic strengths), the bacterial cells is found unable to adhere on the surface. In contrast, at high ionic strength, the energy disappears and bacterial cells can easily and rapidly attain irreversible adhesion.

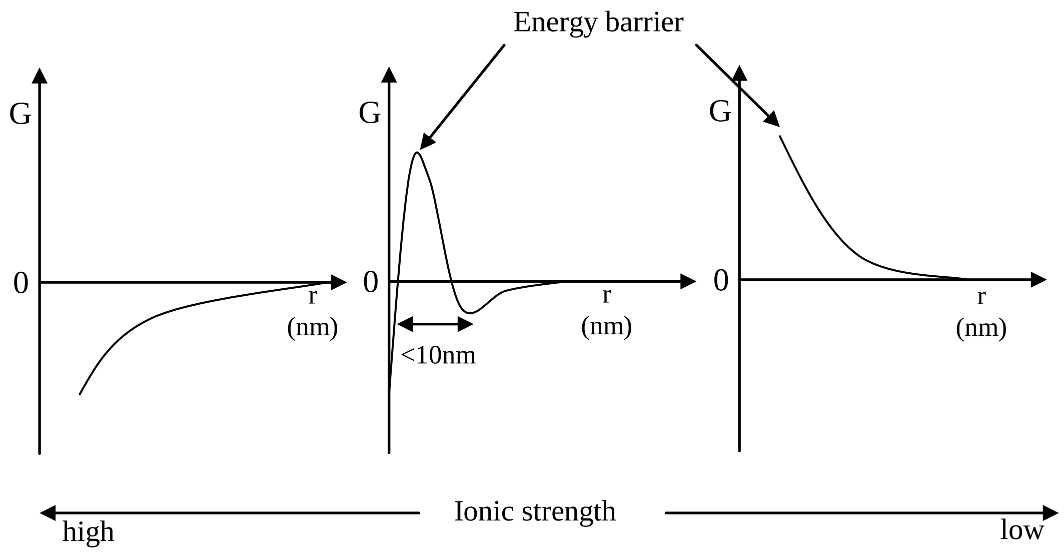

Figure 2.5: Total interaction energy between a bacterial cell and a surface depending on ionic strength (Hori and Matsumoto, 2010).

The retention of bacteria via adsorption mechanism is influenced by porous media designation (Huysman and Verstraete, 1993). The smaller the particle sizes, the larger the surface area, hence more adhesion sites will be provided (DeFlaun and Mayer, 1983; McDowell-Boyer, 1986).The surface roughness of the porous media may increase the adsorption (as a result of reduced sheer forces) and lower desorption rates, thus increase the media surface area (Donlan, 2002).

2.6 Bacterial adhesion and biofouling

Water filtration based on membrane technology is frequently coupled with an undesired decline in flux which is caused by membrane fouling. Fouling is generally defined as a reduction in water transport per unit area of membrane caused by the accumulation of substances including microorganisms, inorganic, particulates, colloidal and organic matter on or in the membrane (Lee *et al.*, 2010). Biofouling during bacteria removal may cause significant effect to osmotic pressure and hence require frequent chemical cleaning which eventually shorten membrane life.

The adhesion/interaction between particles, including both inorganic colloidal particles and bacterial cells has led to biofilm formation. The particles adhered on or in the membrane form biofilm and reduce the flow through the membrane, which in turn result in great reduction in the filtration efficiency and working lifetime of the membranes (Hilal *et al*., 2009). Biofilms that composed primarily of microorganisms and extracellular polymeric substances is the major hindrance in membrane filtration and cause severe loss of performance.

A study of Lee *et al*. (2010) which explored the PES UF membrane (350 Da) and polyamide (PA) NF membrane performances on *Staphylococcus epidermis* (*S.epidermis*) (0.5 μm, gram positive, sphere), *Flavobacterium lutescens* (*F.lutescens*) (2.5 μm × 0.3μm, gram negative, rod) and *Escherichia coli* (*E.coli*) (1.5 μm × 0.5 μm, gram negative, rod) has resulted in few conclusions:

i) In terms of particles retention (under a high ionic strength condition), the PA NF membrane exhibited a much lower fouling than that of PES UF membrane.
ii) PES UF membrane which is rougher and more hydrophilic surface showed lower retention time in which the lower the retention time, the lower propensity for membrane biofouling.
iii) Bacteria retention on membrane surfaces were longer in KCl solution (stronger ionic) compared to deionized water.
iv) Among the three bacteria sp., *E.coli* and *F.lutescens* exhibited the highest potential of fouling for both membranes.

Kochkodan *et al*. (2008) studied the adhesion of different microorganisms to polymeric membrane of various chemical natures. Results revealed that membranes deposited with TiO_2 particles reduced the number of cells in colony forming unit per mL (CFU/mL) about 98.1 % under uv-irradiation for six hours. The mechanism of bactericidal action of TiO_2 under black uv-irradiation is based on the formation of OH, O_2^- and HO_2 radicals in aqueous system (Salih, 2002). It was also found that microorganisms adhered more on hydrophobic PES or PSf than on hydrophilic cellulose membrane. Hydrophilic *E.coli* was found to adhere less compared to hydrophobic *P.putida*. In addition, the fluxes of membrane deposited with microorganisms was found to decrease with time and TiO_2 particles presence has provided a strong photo bactericidal under black uv-irradiation.

According to Hori and Matsumoto (2010), the bacterial adhesion can be controlled by antimicrobial agents' addition, surface modifications or electro-classical methods.

2.7 Advantages of antibacterial membrane over the other bacteria removal method

Conventional membranes have optimized pore sizes and other membrane properties such as hydrophilicity to remove bacteria. The key advantage of an antibacterial membrane is the enhanced antibacterial action by the incorporated antibacterial agent. Permeability on the inner and outer antibacterial membranes will lead to the release of antibacterial agent and ultimately disrupt the bacterial cell wall membrane. Therefore, instead of bacteria removal via retaining them on membrane surfaces, an antibacterial membrane offers removal (via suitable pore size ranges) and disinfection in a standalone system.

As a result, biofilm formation can be substantially hindered and biofouling can be obstructed. The formation of smoother and anti-adherence membrane surfaces is another value added which then extend an antibacterial membrane as a promising candidate in bacteria removal for wastewater treatment. It has been proposed that the interaction energy between a colloidal particle and a rough membrane surface has considerable lateral variations thus particles will have greater tendency to accumulate (Rizwan and Bhattacharjee, 2007). In contrast, a smooth surface reduces or eliminates any non-contacting areas thus increases the repulsive interaction energy barrier between a colloidal particle and membranes surfaces (Hoek *et al*., 2003).

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