

Thesis Approval

Microbial Removal From wastewater Using Membranes Technology

Prepared By: Rinad Mouas Saleh

Registration No: 20611025

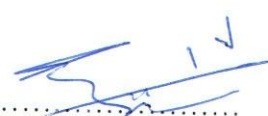
Supervisor: Dr Jihad Abbadi

Co- Supervisor: Prof. Mustafa Khamis

Master thesis Submitted and accepted, 23rd of January 2011. Names of examiners
commute members are as following:

1- Head of Committee

Dr. Jehad Abbadi

Signature.....

2- Internal Examiner

Dr. Mutaz Ali. Qutob

Signature

3- External Examiner

Dr. Marwan Ghanem

Signature

Abstract

Palestinian territories are suffering from water shortage. High population growth generate a large amount of wastewater with sanitation problems, which causes diseases related to the microbial contamination in drinking water, so the demand for clean water is growing. To solve these problems a lot of wastewater treatment plants were established in Palestine, most of them are based on biological treatment such as aeration bonds, wet land, and activated sludge process. Membrane technology has been widely spread in many countries in the past decade, for both water purification and wastewater treatment for their efficiency in removing bacteria and viruses.

Most recently, the combination of membranes with the activated sludge process has redefined basic sewage treatment, optimizing the biological treatment operation and yielding a treated effluent that is ideal for the reuse bringing treated wastewater to four star grade according to the revised WHO standards. Membranes are available in a diverse range of forms and configurations, with membrane pore size, or molecular weight (MW) separation being the primary characteristic of differentiation. Based on this type of differentiation, systems are classified as microfiltration, ultra-filtration or reverse osmosis.

Our study based on testing the efficiency of a consequent system of wastewater treatment consisted of: activated sludge process followed by a hollow fiber ultra-filtration membrane system followed by a spiral wund ultra-filtration membrane system followed by a reverse osmosis system in the removal of different types of bacteria in terms of their cell wall structure (gram stain) without the usage of chlorination as a disinfectant to avoid its health and environmental bad complications.

This study was conducted in the wastewater treatment plant at Al-Quds University in Abu-Dies - Jerusalem/Palestine. The wastewater was collected from Al-Quds university campus. The wastewater treatment plant is consisted of an activated sludge process followed by hollow fiber membranes ultra-filtration system (with 100 kD cutoff), followed by a spiral wound membrane ultra-filtration system (has pore size range from 0.005 to 0.04 μ m), followed by a reverse osmosis system (can remove molecular weight greater than 150-250 Daltons).

This study was based on firstly screening the wastewater generated from the university campus in terms of its microbial characteristics and the efficiency of the designed plant in the removal of screened bacteria, secondly passing different types of bacteria differ in

terms of their cell wall structure (gram stain characteristic) in specific count in a modeling experiment in order to test the efficiency of the designed treatment plant in removing model bacteria based on their gram stain characteristic. The research was conducted in the period from July 2008 till January 2010, investigating some species of bacteria that has certain health significance. The microbes that were screened were: *Total coliform*, *Fecal coliform*, *Total microbial count*, and other species such as *Escherichia coli*, *Enterococci spp.*, *Shigella* and *Salmonella*. The last four species that are mentioned above were used as model bacteria in the modeling experiment and were grown in the laboratory under complete sterile technique to a specific count, and were separately inoculated in a 1500L tank, which was filled with 500 L fresh water. The inoculated bacteria were passed through the consequently ordered membrane systems of the treatment plant. The count of bacteria under investigation was tested in the effluent of each type of filters and in its brine.

The screening part of the research represented that the hollow fiber UF membrane was efficient to reject 90% of the Total microbial count (TPC), 90% of the Total coliforms (TC), and also 90% of the Fecal coliforms (FC). It showed no significant efficiency in the rejection of *E. coli* and *Enterococcus spp.* While spiral wound showed a high efficiency in the removal of microbes under study by 2-3 log (99 - 99.9%) for TPC, and was efficient to eliminate TC, and FC completely. However, RO effluent was free from these microbes. *Salmonella* and *Shigella* bacteria were not present in the wastewater effluent that entered the treatment plant so the efficiency of their rejection was not checked in the screening part.

The modeling experiment revealed that the hollow fiber was efficient to remove 99% of the inoculated *E. coli* (gram negative bacteria), and 99-99.9% of *Enterococcus spp.* (gram positive bacteria). While its efficiency in the removal of *Salmonella* and *Shigella* (gram negative bacteria) reached to 90-99% of the effluent's bacterial count. The spiral wound was more efficient to reject 99.9% of *E. coli*, 99.999%, of *Enterococcus spp.*, 99.99% of *Salmonella*, and 90-99.9% of *Shigella*. The reverse osmosis effluent was free from microbes, which is considered as the most efficient membrane in the rejection of entered bacteria under study.

Our results showed that, there was no significant difference between gram negative and gram positive type bacteria in terms of their forward fouling (they pass through the membranes that have pores much smaller than the microbe that leak from the membrane). This was discussed not in terms of their cell wall structure but could be due to

imperfections in the membrane. These imperfections could be due to the construction of the membrane, membrane degradation, broken fibers, and scratches in the membrane surface that lead to the formation of pores larger than the theoretical size. Based upon this work we concluded that the microbes disinfection can take place by activated sludge process, followed by ultrafiltration - hollow fiber, and spiral wound – which they were highly efficient to reject most of the TPC, TC, FC, E.coli, Enterococcus spp. Salmonella, and Shigella to a satisfactory removal without chlorine usage, but reverse osmosis membrane eliminated all these types completely without facing problems of microbial fouling or cake formation that suppress filtration. Therefore we encourage municipalities to use these integrated system to treat wastewater for industry and agriculture reuse, and put education and training strategies for persons they involved with onsite wastewater system, and also encourage other researchers to study viruses in water and wastewater using these types of membrane technology.

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Chapter One

I Introduction and Literature Review

1.1 Water Status in Palestine

The Mediterranean Region is suffering from significant water shortage and the demand on clean water is increasing. As a result, the search for new resources along with protecting used ones becomes a very pronounced priority for the researchers and the decision makers in the area in order to preserve water resources and protect its quality (Al-Sa'ed, 2000). The main sources of fresh water in Palestine are the Jordan surface and the ground water underlying the west bank and the coastal area. As Israel denies the Palestinian rights to share and use water from Jordan River, only 10 % of the ground water resources is allocated to the Palestinians (Palestinian Media center, 2003). Therefore, the Palestinian water Authority (PWA) was established after Oslo agreement (1994) to monitor, develop and distribute the limited drinking water supply allocated to it by the Israeli's authority (UNEP, 2003). The unfair distribution of water resources and the denials of the full control of the Palestinians over their natural water reservoirs make most of the Palestinian municipalities and villages to suffer severely from shortages in fresh water supply especially in summer. These cities and villages are compelled to implement rotation plans, particularly during summer, to distribute the little water available. Under these plans, residents in a particular area of the city receive water for a day. The flow is shut off for several days, while water is supplied to other areas until the sector's turn comes again. Hebron, Bethlehem, Jenin, Ramallah and some Palestinian neighborhood in Jerusalem implement such plans. All major villages are facing even more serious problems (UNEP, 2003). Also specific daily water consumption is low with less quality of water, and this in turn will reflect the situation of the overall economy. Actually, getting water easily and available all the time in good quality is considered to be fundamental human rights. (Al-Sa'ed, 2000).

Additionally, the demand on water is growing rapidly in Palestine due to rapid population growth, urbanization and socioeconomic development. Consequently generated wastewater in both the West Bank and Gaza Strip is increasing in volume, making these areas facing a series of wastewater and sanitation-related problems. These problems including; large-scale discharge of untreated wastewater, leaking of collected wastewater from sewer systems and cesspits, water treatment plants that do not function or function

only poorly, and uncontrolled reuse of untreated wastewater by the irrigation sector . Accordingly, wastewater related problems cause many diseases related to contaminated drinking water. Statistics from the Center for Disease Control and Prevention (CDC, 2003) reveal that there were more than 17,000 cases of water-related illnesses during recent two years in Gaza Strip (Table1.1 and Table1.2). However, since many symptoms are often confused with other sicknesses, some researchers feel as many as 25 outbreaks go unreported for every one reported, since many people are exposed to potentially harmful microbes and pesticides, through drinking tap water and taking showers (UNEP, 2003).

The major source of waterborne diseases in Gaza is the direct or indirect contamination of drinking water by raw or ineffectively treated wastewater, as consequent poor treatment allows the coliform bacteria to persist. Although, most wells and networks in the Gaza Strip were found to be free from organisms (Ministry of Health), however, some pathogens will survive for only a short time in water, having small chance to cause illness, but others may survive for months like *Giardia*, *Amoeba* and *Cryptosporidium*. If water is not treated or poorly treated for pathogens, a susceptible person will suffer from illness (WHO, 2003).

Table 1.1: Tested water samples for coliform bacteria in different Governorate in Gaza Strip (WHO, 2003)

Governorate	No. of samples tested	No. of contaminated samples	Contaminated %
North	749	108	14.4
Gaza City	2907	477	16.4
Mid-Zone	714	103	14.4
Khan Younis	350	74	21.1
Rafah	385	42	10.9
Total	5105	804	15.7

As reported by the Palestinian Ministry of Health (MOH, 2002), it was shown that, of 4915 samples collected from water networks and wells in Gaza, 638 (12.8% of the samples) were contaminated with microbes originated from wastewater (Coliform bacteria). Additionally, in 2003, out of a total 5105 samples collected from wells and networks in Gaza Strip, 804 samples were found to be contaminated which means that, the instances of contamination increased to 15.7% compared to 12.8% in 2002. The World

Health Organization recommends that the percentage of coliform contamination in water samples from water wells and networks should not exceed 4% (MOH, 2002). Pathogens and diseases spread by the fecal-oral route (water borne diseases), and can be caused by bacteria (salmoellosis, typhoid fever and cholera), viruses (viral gastroenteritis and hepatitis-A and E) and protozoa (Guardiasis, amoebic dysentery and Cryptosporidiosis) (Table 1.2). Many people who live in Beit Lahia suffer from chronic gastrointestinal diseases and meningitis with increased infections in summer as a direct result of more people swimming in the beach camp shore (Randa El Khodary, 2003).

Table 1.2 Waterborne pathogens and their associated illnesses (Melad, 2002)

Organisms	Associated health effect
1. Viruses	
<i>Hepatitis A</i>	Fever, nausea, jaundice, liver failure
<i>Hepatitis E</i>	Fever, nausea, jaundice, death diarrhea
2. Bacteria	
<i>Escherichia coil</i>	Gastroenteritis (diarrhea)
<i>Salmonella sp</i>	Enterocolitis (fever, diarrhea, vomiting)
<i>Shigella sp</i>	Gastroenteritis, reactive arthritis
<i>Vibrio Cholera</i>	Diarrhea, vomiting, death
3. Protozoa	
<i>Cryptosporidium parvum</i>	Diarrhea
<i>Giardia lamblia</i>	Chronic diarrhea

1.2 Wastewater

Wastewater is the water discarded after it has been used for domestic, commercial or industrial purposes, usually requires treatment before discharge, in order to protect the environment (Barnes et al., 1981). Wastewater is characterized in terms of its physical, chemical, and biological composition. Raw wastewater originates from domestic and industrial sources. The characteristics of wastewater are impacted by water consumption rates, population density, industrial practices, and habits of the population. The discharge of raw wastewater causes a major potential health hazard as it carries disease in the form of pathogens and toxic chemicals. Disposed raw wastewater mixes with seasonal surface

water, and part of it percolates into the ground and finally may affect groundwater quality (Metcalf and Eddy, 1991).

1.3 The important contaminants of concern in wastewater

Total solids: are defined as all the matter that remains as residue upon evaporation at 103 to 105°C. Matter that has been significant vapor pressure at this temperature is lost during evaporation and is not defined as a solid. Settleable solids are those solids that will settle to the bottom of a cone-shaped container (called an Imhoff cone) in a 60 minute period, expressed as ml/L, are an approximate measure of the quantity of sludge that will be removed by primary sedimentation. Total solids or residue upon evaporation can be classified as nonfilterable (suspended) or filterable by passing a known volume of liquid through a filter. The filterable- solids fraction consists of colloidal and dissolved solids with size range from 0.001 to 1µm. The dissolved solids consist of both organic and inorganic molecules and ions that are present in true solution in water. The colloidal fraction cannot be removed by settling, but could be removed by biological oxidation or coagulation, followed by sedimentation. Each of categories of solids may be further classified on the basis of their volatility at 550 ± 50°C. The organic fraction will be oxidized and will be driven off as gas at this temperature, where the inorganic fraction remains behind as ash. Therefore, volatile suspended solids and fixed suspended solids, refer to the organic and inorganic (or mineral) content of the suspended solids respectively (Metcalf & Eddy, 1991).

Biodegradable organics: These are composed principally of proteins, carbohydrates and fats. They are measured most commonly in terms of BOD (biochemical oxygen demand) and COD (chemical oxygen demand). If untreated wastewater is discharged to the environment, the biological stabilization of the biodegradable organics can lead to the depletion of natural oxygen resources and development of septic conditions (Metcalf and Eddy, 1991).

Pathogens: Waterborne diseases can be transmitted to people by the pathogenic organism in wastewater.

Chapter five

V. Conclusions and recommendations

5.1 Conclusions

Based upon this work and the experiments done, the following points can be concluded.

- Activated sludge process reduced the initial numbers of TPC, TC, and FC to about 90-99%
- UF- hollow fiber membrane was efficient to reject about 90% from TPC, TC, and FC, and about 99% of *E. coli*, 99-99.9% rejection of *Enterococcus spp.*, and 90-99% rejection of *Salmonella* and *Shigella*
- Spiral wound was more efficient to remove about 99.9% from TPC, and eliminate TC, FC completely, also 99.9% of *E. coli*, 99.999% of *Enterococcus spp.*, 99.99% of *Salmonella*, and 90-99.9% of *Shigella*,
- RO membrane was efficient to remove all the microbes under investigation completely.
- The pretreatment process and activated sludge followed by hollow fiber ultra-filtration followed by spiral wound ultra-filtration was highly efficient to reject most of the TPC, TC, FC, *E. coli*, *Enterococcus spp.*, *Salmonella*, and *Shigella* moreover RO membranes eliminate all these types of bacteria completely without facing problems of microbial fouling (cake formation that suppress filtration)
- The chlorination process in some cases after activated sludge process was not efficient to kill all bacteria, so using membrane technology method reduced the chlorine usage as disinfectant, since chlorine has bad environmental impacts.
- Disinfection can take place without the need for chemicals which, could be achieved using ultra-filtration and reverse osmosis membranes.
- Membrane technology depends on pore size and their distribution within a membrane is very important in the potential of the membrane in microorganism's removal.
- UF Processes with pore size approximately 0.01 μm and smaller, and RO membranes are considered as absolute barriers for many pathogens of concern to the water treatment industry, but could be subjected to leak of some microbes.

- There was passage of some pathogens and bacterial through membrane pores, which was observed in the membranes permeate. There are parathion and operational concerns that should be addressed.
- Imperfections in the construction of the membrane module or degradation of the membrane system over time can lead to the passage of microorganisms into the treated water. These imperfections can include broken fibers, scratches in the membrane Pores surface, which are larger than the nominal size, membranes that do not seal properly, and glue joints that may be cracked.

5.2 Recommendations

- It should be noted that wastewater treatment has three stages, every one depends on each other to remove bacterial indicators and pathogens completely, the first is activated sludge followed by ultra filtration (hollow fiber and spiral wound), followed by reverse osmosis. The work is consequence and if one stage does not exist then membrane fouling and inefficient treatment will occur, so the work in this order and stage by stage is recommended.
- Encouraging other researchers to study viral contamination in fresh and wastewater using these types of technological membranes.
- Encouraging other researchers to take (brine) which is membrane rejection as case study in future.
- It is recommended that the municipalities should construct the wastewater treatment plant specially in agricultural areas using membrane technology instead of traditional wastewater treatment pools, because it is efficient and don't need for a large spaces
- It is recommended that, people who are working with local governments and other stakeholders should develop strategies and programs for education and training of persons involved with onsite wastewater system use, regulation, design, installation, maintenance or inspection.

ملخص العربية

إزالة المايكروبات من مياه الصرف الصحي باستخدام تقنية الأغشية
الاسم: ريناد موسى صالح

تعاني المناطق الفلسطينية من نقص في مياه الشرب، وذلك بسبب النمو السكاني الذي ينتج كمّاً هائلاً من مياه الصرف الصحي، و التي تعد مشكلة على الصحة العامة؛ بسبب محتواها الميكروبي، الذي يسبب أمراضاً خطيرة للإنسان مما أدى إلى زيادة الطلب على المياه الصالحة للشرب. ولحل هذه المشكلة انتشرت العديد من محطات التنقية في فلسطين معظمها يعتمد على المعالجة البيولوجية مثل برك التنقية، الأراضي الرطبة، الحمأة المنشطة.

إن تقدم تكنولوجيا الأغشية في العقد الماضي، أدى إلى تطور عملية التنقية و معالجة مياه الصرف الصحي لكفاءتها في إزالة البكتيريا و الفيروسات. وفي الآونة الأخيرة تم استخدام الحمأة المنشطة، بالإضافة إلى الأغشية؛ لتحسين عملية المعالجة التي تضمن إعادة استخدام مياه الصرف الصحي المعالجة. وقد توفرت الأغشية بتراكيب و أشكال متنوعة يتم التمييز بينها بواسطة حجم مسام الغشاء أو الوزن الجزيئي للأجسام التي تستطيع إزالتها. واستناداً إلى هذا النوع من التمايز تم تصنيف الأغشية إلى الترشيح الدقيق Hollow fiber، الترشيح الفائق Spiral wound، يليها نظام التناضح العكسي Reverse osmosis .

واستخدمت هذه الأغشية في محطة جامعة القدس/أبو ديس /فلسطين. لمعالجة مياه الصرف الصحي التي تجمع من الحرم الجامعي، حيث يتم ضخها إلى محطة المعالجة التي تتكون من الحمأة المنشطة، يتبعها نظام الأغشية المكون من الأغشية المجوفة Hollow fiber التي تصل قدرتها إلى إزالة الجزيئات التي يزيد حجمها عن 100 كيلو دالتون. يليها الأغشية الفائقة الترشيح Spiral wound التي تتراوح مسامها بين 0.04-0.055 ميكرون. يتبعها غشاء التناضح العكسي Reverse osmosis ذو القدرة على إزالة الجزيئات التي يزيد وزنها الجزيئي عن 150-250 دالتون. و قد قام البحث على فرضية تفيد إمكانية مرور البكتيريا عبر الأغشية رغم صغر مسامها إلى المياه المعالجة، تبعاً لنوع الجدار الخلوي المحيط بالخلية سواء كانت البكتيريا سالبة جرام والتي يتميز جدارها بالمرونة، أو موجبة جرام ذات الجدار الصلب، أو ربما التسرب نتيجة لوجود خلل في صناعة الأغشية. حيث استند البحث على الأهداف التالية:

أولاً : تقليل الأثر السلبي لمياه الصرف الصحي الناتج من استخدام المواد الكيميائية، خصوصاً الكلور في تطهير المياه المعالجة، و زيادة كفاءة معالجتها باستخدام تقنية الأغشية.

ثانياً: تقييم كفاءة هذه الأغشية لإزالة البكتيريا و الجراثيم و ذلك بتصميم تجربة يتم من خلالها حقن أنواع مختلفة من البكتيريا في خزان، و من ثم تمريرها خلال أغشية الألياف المجوفة Hollow fiber، يتبعها الأغشية فائقة الترشيح Spiral wound، يليها غشاء التناضح العكسي Reverse osmosis.

ولتحقيق الأهداف تم البحث على جزأين الأول: امتد نحو عامين من (كانون ثاني سنة 2008- إلى تموز سنة 2010) وامتازت هذه المرحلة (Screening) بفرز أنواع معينة من البكتيريا مثل *Enterococcus* ، *E. coli* ، *Fecal coliform* ، *Total coliform* ، *Total plate count* ، *Salmonella* ، *Shigella* ، *spp.* و قد استخدمت الأنواع الأربعة الأخيرة كنموذج تجربة للتحقق من فعالية الأغشية في التخلص منها، حيث تم تنميتها بشكل منفصل في المختبر تحت ظروف معقمة إلى تراكيز معروفة، تعطي توقع لأعداد البكتيريا التي سيتم حقنها بشكل منفصل (كل نوع بكتيريا على حدى) في خزان سعته 1500 لتر معبأ ب 500 لتر من المياه العذبة. و من ثم يتم إدخالها إلى محطة الأغشية (Hollow fiber, Spiral wound) يليها غشاء Reverse osmosis.

حيث أشارت نتائج مرحلة التخليق Screening إلى أن فعالية غشاء الألياف المجوفة Hollow fiber كان ذو قدرة على إزالة 90% من *TPC* , *FC* , *TC* و لم يظهر فعالية بشكل واضح في إزالة *Enterococcus* , *E.coli*.

بينما أظهر غشاء الترشيح الفائق Spiral wound فعالية في إزالة 90-99.9% من *TPC* و إنهاء وجود *FC* , *TC* فكانت المياه الداخلة لغشاء التناضح العكسي Reverse osmosis تخلو من هذه الميكروبات. كما أظهرت النتائج خلو مجتمع الدراسة (جامعة القدس) من *Salmonella* , *Shigella* ,

أما بالنسبة لنتائج التجربة (النموذج) فقد أظهرت فعالية الغشاء المجوف Hollow fiber في إزالة 99% من *E. coli* (سالبية غرام)، 99-99.9% من *Enterococcus spp.* (موجبة غرام) وإزالة ما يقارب 90-99% من *Salmonella* , *Shigella* (سالبية غرام).

بينما أشارت نتائج الغشاء الفائق الترشيح Spiral wound بأنه أكثر فعالية من الغشاء السابق، حيث نجح في إزالة 99.9% من *E.coli* ، 99.9-99% من *Enterococcus* ، 99.99% من *Salmonella* و 90-99.9% من *Shigella*. أما غشاء التناضح العكسي Reverse osmosis كان الأكثر فعالية بين الأغشية في إزالة هذه الميكروبات نهائياً، لكن مع الوقت يلاحظ تسرب الميكروبات من خلال الأغشية إلى المياه المعالجة وذلك يعزى إلى خلل في تركيب هذه الأغشية أو خدش في سطوحها مما يؤدي إلى توسع تقوُّبها، وهذا ينفي فرضية مرور الميكروبات خلال الأغشية بسبب نوع الجدار الخلوي (سالبية غرام أو موجبة غرام).

وبذلك نستنتج إن إنشاء محطة معالجة مبنية على عملية الحمأة المنشطة يليها نظام متكامل من الأغشية سواء كانت الأغشية المجوفة، أغشية الترشيح الفائق، والتناضح العكسي تكون فعالة في التخلص من الميكروبات نهائياً دون اللجوء إلى الكلور كمطهر، ولذلك أدعو المجالس البلدية إلى إقامة محطات لمعالجة المياه تعتمد على نظام متكامل يشمل تقنية الأغشية لإعادة استخدام المياه المعالجة في الصناعة و الزراعة وإقامة إستراتيجية لتعليم و تدريب أشخاص للعمل في تلك المحطات، كما أشجع الباحثين إلى استخدام مثل هذه المحطات لعمل دراسة على فعاليتها في التخلص من الفيروسات.