OPTIMIZATION OF POLYETHERSULFONE ULTRAFILTRATION HOLLOW FIBER MEMBRANE FOULING PERFORMANCES USING PARTICLE SWARM OPTIMIZATION

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DEDICATION

This thesis is dedicated to my father, who taught me that the best kind of knowledge to have is that which is learned for its own sake. It is also dedicated to my mother, who taught me that even the largest task can be accomplished if it is done one step at a time.

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ABSTRACT

Nowadays, membrane separation has become a popular method for various industries worldwide. Membrane separations are often used in water filtration, food industries and gas separation. For more efficient use of membranes, the control of membrane fouling, which can be defined as fouling requiring reagents is of importance. Fouling of membranes is a significant issue for the efficiency of membrane filtration in wastewater treatment systems. Research on ultrafiltration membranes for water treatment is gaining more attention especially in production sectors. However, in solving the reduction of fouling condition problems, previous studies mostly used an experimentation that varied one of the independent filtration conditions and fixed the others. The common problem is the ultrafiltration process cannot be performed effectively due to non-optimum settings of the filtration conditions. Hence, in order to solve these issues, this study aims to use Particle Swarm Optimization (PSO) to optimize the polyethersulfone (PES) ultrafiltration hollow fiber membrane conditions for oily wastewater treatment to maximize fouling index. In this experiment, five variables were evaluated. They were pH and temperature of feed solution, time, transmembrane pressure and surface area of membrane. In order to minimize the number of experiments but still capable of quantifying the effect of each variables, Response Surface Methodology (RSM) of half factorial design was applied. The experimental plan was based on a combination of high levels and low levels, half factorial designs with resolution V, center points, as well as axial points. Furthermore, the regression models were generated by employing the Design Expert 6.0.5 software and they were found to be significant and valid. Then, the regression models obtained were proposed as the objective functions of PSO to determine the optimal fouling conditions. The MATLAB software was used to code and execute the PSO. Based on the results, the optimal conditions occurred at pH of 11.40, temperature of 32.5 °C, time of 28 minutes, transmembrane pressure of 2.97 bar and surface area of 0.042 m^2. The membrane morphology under the influence of different ultrafiltration conditions was investigated via scanning electron microscope (SEM). As a conclusion, the fouling index during the ultrafiltration process of PES hollow fiber membrane has been optimized to reduce membrane fouling. The experimental results of this study can help to reduce the fouling of membranes, thus contributing to a more sustainable filtration system. As a future research direction, the solutions from PSO can be compared with other optimization techniques such as Genetic Algorithm (GA).

ABSTRAK

Pada masa kini, pemisahan membran semakin dikenali di pelbagai industri di dunia. Pemisahan membran sering digunakan di dalam penapisan air, industri makanan dan pemisahan gas. Untuk penggunaan membran yang lebih cekap, kotoran membran hendaklah dikawal dengan mengenal pasti reagen pengotoran adalah sangat penting. Kotoran membran menjadi isu yang utama bagi kecekapan proses penapisan membran di dalam sistem rawatan sisa air. Penyelidikan ultrapenurasan membran untuk rawatan air amat popular terutamanya di dalam sektor pengeluaran. Walaubagaimanapun, di dalam mengurangkan masalah pengotoran, kebanyakannya kajian lepas ditangani dengan menggunakan eksperimen yang hanya mengubah salah satu keadaan semasa proses penapisan dan menetapkan keadaan yang lain. Masalah yang biasa berlaku ialah proses ultrapenurasan yang tidak dapat dilakukan dengan berkesan kerana keadaan tetapan yang tidak sesuai. Oleh itu, untuk menyelesaikan isu ini, tujuan kajian ini adalah dengan menggunakan kaedah pengoptimuman kumpulan zarah (PSO) untuk mengoptimumkan keadaan membran gentian geronggang ultrapenurasan polyetehersulfone (PES) yang digunakan dalam rawatan air sisa berminyak untuk meningkatkan indeks pengotoran. Dalam ekperimen ini, lima pembolehubah telah dinilai. Ia adalah pH, suhu cecair kajian, masa, tekanan transmembran dan luas permukaan membran. Untuk mengurangkan bilangan kajian tetapi masih berkeupayaan untuk mengukur kesan setiap pembolehubah, Metodologi Permukaan Tindakbalas (RSM) dengan reka bentuk faktoran pecahan telah digunakan. Pelan eksperimen ini berdasarkan gabungan tahap tinggi dan tahap rendah, reka bentuk faktoran separuh dengan resolusi V, titik tengah dan mata paksi. Perisian Design Expert 6.0.5 telah menghasilkan model regresi dan model didapati penting dan sah. Kemudian, model regresi yang diperolehi dicadangkan sebagai fungsi objektif PSO untuk menentukan keadaan pengumpulan kotoran optimum. Perisian MATLAB digunakan untuk mengaturcara dan melaksanakan PSO. Berdasarkan keputusan, kedudukan keadaan optimum berlaku apabila pH adalah 11.40, suhu adalah 32.5°C, masa adalah 28 minit, tekanan transmembran adalah 2.97 bar dan luas permukaan membran adalah 0.042 m^2. Morfologi membran di bawah pengaruh keadaan ultrapenurasan berbeza disiasat menggunakan mikroskop imbasan electron (SEM). Sebagai kesimpulan, indeks pengotoran semasa proses ultrapenurasan membran gentian beronggang PES telah dioptimumkan untuk mengurangkan pencemaran membran. Hasil kajian ekperimen ini dapat membantu mengurangkan kotoran membran sekaligus menyumbang kepada sistem penapisan yang lebih lestari. Untuk penyelidikan masa hadapan, penyelesaian daripada PSO boleh dibandingkan dengan teknik pengoptimuman yang lain seperti Genetik Algoritma (GA).

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

LIST OF ABBREVIATIONS

AI	-	Artificial Intelligence
ACO	-	Ant Colony Optimization
AMTEC	-	Advanced Membrane Technology Center
ANN	-	Artificial Neural Network
ANOVA	-	Analysis of Variance
BFT	-	Bore Fluid Temperature
BFR	-	Bore Fluid Ratio
CA	-	Cellulose Acetate
CAB	-	Cellulose Acetate Butryrate
CAP	-	Cellulose Acetate Propionate
CBT	-	Coagulation Bath Temperature
CIP	-	Cleaning in Place
CN	-	Cellulose Nitrate
CP	-	Cellulose Propionate
D/DBP	-	Disinfectants/Disinfection by Product
DER	-	Dope Extrusion Rate
DOC	-	Dissolved Organic Compound
DOE	-	Design of Experiment
EC	-	Ethyl Cellulose
EfOM	-	Effluent Organic Matter
FA	-	Fulvic Acids
FI	-	Fouling Index
FL	-	Fuzzy Logic
GA	-	Genetic Algorithm
GP	-	Genetic Programming
HA	-	Humic Acids
H_2O	-	Water
HFM	-	Hollow Fiber Membrane
MATLAB	-	Matrix Laboratory
MF	-	Microfiltration

ML	-	Machine Learning
MLPANN	-	Multilayer Perception Artificial Neural Network
MT	-	Model Trees
MWCO	-	Molecular Weight Cut-off
NF	-	Nanofiltration
NMP	-	*N*-methyl-2-pyrrolidone
PA	-	Polyamide
PC	-	Polycarbonate
PE	-	Polyethylene
PEG	-	Poly(ethylene) Glycol
PES	-	Polyethersulfone
PP	-	Polypropylene
PRESS	-	Presicted Residual Sum of Squares
PSO	-	Particle Swarm Optimization
PU	-	Polyurethane
PVDF	-	Polyvinylidene Fluoride
RO	-	Reverse Osmosis
RSM	-	Response surface methodology
SDI	-	Silt Density Index
SDWA	-	Safe Drinking Water Act
SEM	-	Scanning Electron Microscope
SMBR	-	Submerged Membrane Bio-Reactor
SS	-	Spinneret size
SWTR	-	Surface Water Treatment Rule
TM	-	Taguchi Method
TMP	-	Transmembrane Pressure
UF	-	Ultrafiltration
UTM	-	Universiti Teknologi Malaysia

LIST OF SYMBOLS

A	-	Surface Area
C_1	-	Acceleration constant for the cognitive component
C_f	-	Concentration of solute area of feed
C_p	-	Concentration of solute area of permeate
C_2	-	Acceleration constant for the social component
d	-	Dimension being considered, each particle has a position and velocity for each dimension
H	-	pH
i	-	Particle's index, used as a particle identifier
it	-	Iteration number, the algorithm is iterative
g_{bd}	-	The location in dimension d with the best fitness among All the visited locations in that dimension of all the particles
P	-	Transmembrane pressure
$pb_{i,d}$	-	The location in dimension d with the best fitness of all The visited locations in that dimension of particle *i*
R	-	Rejection
R_{nd}	-	Stochastic component of the algorithm, a random value between 0 and 1
t	-	Time
T	-	Temperature
$v_{i,d}$	-	Velocity of particle *i* in dimension *d*
$x_{i,d}$	-	Position of particle *i* in dimension *d*

LIST OF APPENDICES

CHAPTER 1

INTRODUCTION

1.1 Problem Background

Membrane separation has become very popular separation in various industries in the world especially in the global oil industry. Membrane separation is often used in water filtration, food industry and gas separation. Nowadays, global oil demand is increasing due to the rapid development of many industries, such as high vehicle and fuel consumption for the manufacturing industry. By the year 2015, the global oil demand is expected to increase to 94 million barrel per day (MB/d) (1 barrel = 100-200 L). For every barrel of oil produced, three times of barrel of oily wastewater is generated. As a result, large amounts of oily wastewater have been generated from the oil refining industry (Agustin et al., 2008; Ong et al., 2015). To avoid polluting the environment, all wastewater must be treated before being discharged. Malaysia's maximum oil discharge limit is 10 ppm, which is much lower compared to the other countries (International Energy Agency (IEA), 2021). Since oily wastewater consists of various compositions of harmful hydrocarbons, chemical elements and heavy metals before being discharged into water bodies, it needs to be treated properly. However, biological, chemical, and physical treatments are unable to completely separate oil molecules from water, and the process requires a large working area (El-Naas et al., 2009).

Membrane filtration technologies are considered as one of the best option to devoted oily wastewater treatment to meet the stringent local discharged limit and to deal with increasing global oil demand. Membrane separation processes must become more flexible and practical in order to become one of the most effective and demanding methods used to meet demand in a variety of separation-related industries (Gryta et al., 2001). The ability of this membrane technology to separating the composition of various components into two or more products makes it more popular choice to be selected based on its potential and advantages. The research by Loeb and Sourirajan in early 1960s in high-flux asymmetric membranes have started the development of further membrane separation methods. Membrane filtration is not economically realistic in the last thirty years, but with the revolution of new technologies, processes, and material used, membrane technology has been acknowledged as a commercially attractive and highly successful option for purification and separation systems (Wiesner & Chellam, 1999). Therefore, they produce water with stable quality in meeting human, environmental, and industrial demands.

Currently, many membrane separation methods are available. But somehow, there is one of the membrane processes that having rapidly growth in recent years, which is ultrafiltration (UF) processes. Basically, UF membranes is the separation process of very small particles and dissolving macromolecules from compositions using asymmetric membranes with a size of pore between 0.01 to 0.1 μm. Moreover, UF process is the widest method used in many industries compared to other membrane processes because of high efficiency separation technology with low energy consume (Nunes & Peinemann, 2006). The materials used in membrane research include both organic and inorganic materials. Many studies have been conducted in recent years to improve membrane performance in terms of membrane characteristics such as top layer porosity, thickness, sub-layer porosity, and presence of macrovoids, as well as to find new membrane methods and materials for developing high-performance membranes.

In general, membrane performance can be divided into two attributes which are membrane productivity (flux) and extent of separation (rejection of various feed components). Membranes with highest flux and rejection are required, where periodic efforts for maximizing one property will degrade properties of vice versa (Qin et al., 2000). In addition, the process of membrane separation using polymer membranes has been marketed. Polyethersulfone (PES) is selected as the main material (polymer) in this research because of its simple approachability and processing, good selectivity attributes, strong permeability and mechanical characteristics (Li et al., 2004). PES is also identified as an amorphous glassy and hydrophilic polymer in a group of sulphons and is suitable for use in UF separation processes through wet-dry inversion technique.

UF membranes made from PES polymers showed wider level of temperature resistance and different of pH level (Wang et al., 2010).

The phase inversion spinning technique has been generally accepted as the standard technology for manufacturing commercial membranes. It is widely used and has become a popular technology for manufacturing asymmetric hollow fiber membranes. In short, when the spinning solution is immersed and coagulated in the coagulation bath, the phase inversion spinning technique begins. During the whole process, the solvent and non-solvent in the spinning solution are exchanged. As a result, it produces the characteristic structure of an asymmetric membrane, which consists of a dense top layer and porous sub layers (Jung et al., 2004). In this research, asymmetric PES UF hollow fiber membranes would be fabricated according to the dry-wet phase inversion spinning technology.

Membrane modules are another important aspect that needs to be considered because of the approach and performance. Comparing membrane modules, hollow fiber configurations are preferred for industrial practice due to their large membrane packing density, which is used in high membrane areas in small devices (Darvishmanesh et al., 2011). Additionally, compared to flat sheet and spiral wound modules, hollow fiber modules are the favourite option for modules in the filtration method because they have some advantages, high productivity due to their strong mechanical properties, highly flexible modules, and easy handling (Khayet et al., 2012). With these good properties makes hollow fiber membranes very unique from an industrial point of view. Currently, hollow fiber membranes are widely used in more scope of the membrane separation process, such as distillation, UF, nanofiltration (NF), reverse osmosis (RO), and some other filtration processes.

In the area of industrial wastewater treatment, membrane technology has been used to recycle trivalent chromium from tannery wastewater (Fabiani et al., 1996; Shaalan et al., 2001), to remove colour from tannery wastewater (Alves & De Pinho, 2000), to reduce organic polluting compounds in olive-mill wastewater (Turano et al., 2002) and even in artificial kidney mechanisms (Serra et al., 1998). The great usage of UF in industrial operations generates the need for a useful tool for the determination of membrane performance and the minimization of operating costs. The loss of membrane permeability during UF of particles (which is attributed to the adsorption or deposition of particles on the membrane) depends primarily on the interaction of the membrane with the components of the wastewater solution, as well as on the properties of the material of which the membrane has been made. In addition, there are another two contributing factors that should be monitored which are the conditions of the process and the properties of the solution. Therefore, fouling control strategies and sustainable development are very important missions for the community research and technology evaluation programme because water is an important resource for human life. Fouling control strategies are able to decrease energy demand, increase membrane lifetime and reduce other operational costs. Nowadays, modern fouling control approaches focus on changing filtration process variables including alteration of feed water quality (Peiris et al., 2012; Seidel et al., 2002; Busch et al., 2009). Hence, the purpose of this study is to identify any factors that can help reduce the fouling of hollow fiber UF membranes during the separation of wastewater.

1.2 Problem Statement

Research on UF membranes water treatment is hot in the field. With the decrease in material prices, a growing number of membranes used in domestic water treatment provide good results, but membrane fouling is an important obstacle that blocks the promotion of this technology.

For more efficient membrane use, membrane fouling, which can be defined as impurities that require reagents are essential to be treated accordingly. Nevertheless, biological, chemical, and physical treatments cannot fully separate oil molecules from the water and these processes require large areas to be used (El-Naas et al., 2009). Membrane fouling is a major problem that needs to be addressed for the efficiency of membrane filter wastewater treatment systems (Fabris et al., 2007).

Fouling happens when the components are filtered near the membrane or fluid interface. The earliest stages of the fouling process are characterized by concentration polarization (CP) associated with the boundary layer, in which the gradient of the excluded product is formed near the membrane surface (Bader & Veenstra, 1996; Chen et al., 2016). In some cases, excluded products can be made with membrane surfaces or membrane pores, forming what is commonly known as a fouling layer. Some types of fouling layer can be divided into reversible and irreversible fouling based on the strength of the particle connection to the membrane surface.

Membrane fouling is the process where substances or particles dissolve on the surface of the membrane or into the pores of the membrane which indicates membrane performance. This is a main problem that blocks this technology to expand. Membrane fouling will lead to a very poor water quality produced and reduce permeate flux. Dreadful fouling should be cleaned using chemical reaction or membrane replacement. Somehow, this process will increase the cost of treatment process. Loss of membrane permeability during UF (due to deposition or adsorption of particles on the membrane) differs in the interaction of the membrane with the residual components of the wastewater solution, as well as the nature of the membrane material that has been formed. In addition, there are two other contributing factors that must be monitored, which are the properties of the solution and the conditions of the process.

There are limited studies on membrane fouling based on membrane system parameter during filtration process. The related studies of membrane fouling focused on the formulation of membrane fabrication change especially during spinning process parameters (Chung et al., 2000; Chung et al., 2002; Chung et al., 1998; Ismail et al., 2006; Qin et al., 2000; Xu & Qusay, 2004). Besides, Madaeni and Koocheki (2006) explored the parameters of temperature, transmembrane pressure and concentration which affect the flux and rejection in the RO treatment of wastewater containing nitrate, sulfite and phosphate. Meanwhile, Gönder et al., (2010) studied about the effect of pH and temperature during NF process. During the filtration process, there are many parameters that will influence the occurrence of fouling. It must be pointed out that from previous studies in solving these filtration condition optimization problems, they were handled mostly by using an experimentation that involved changing one of the filtration conditions while maintaining the others at fixed levels. For instance, Khan et al. (2016), Ivnitsky et al. (2010), Hesampour et al. (2008a) and

Goosen et al. (2005) varied the transmembrane pressure factor only and fixed other factors in filtration process of hollow fiber membranes. Besides, Lee et al. (2009), Kimura et al. (2004) and Hesampour et al. (2008b) varied the temperature only and fixed other factors during filtration process. Nevertheless, there have been previous studies that used the parameter-by-parameter optimization method to optimize the fouling index (FI) of hollow fiber membranes and it was based on trial and error investigations. This is a very complex process and it requires long time to measure all the parameters for each problem. Furthermore, the complexity of filtration condition problems, as numerous parameters are involved, is one of the main reasons why very little work has been done to vary all these filtration parameters simultaneously. To avoid the time constraints, the parameters that are significant for the fouling occurred must be measured simultaneously. Therefore, there are needs of mathematical modelling to find optimum conditions based on each problem. Even though traditional optimization techniques have the ability of considering several parameters at the same time, they still fail to acquire the relationship equation that links the varied parameters and the outcomes, and besides, it is not easy to discover the optimal parameters combination and optimal response value in the entire area. Taguchi method was applied by Gönder et al. (2010) in order to design the experiments and optimize the experimental results of filtration conditions for cleaning-in-place (CIP) wastewater treatment by NF process while Gönder et al. (2010) used the Taguchi method to get the optimal conditions of pulp and paper mill wastewater treatment using UF process. Besides, Madaeni and Koocheki (2006) also used the Taguchi method in the optimization of wastewater treatment using spiral-wound RO element in their study. However, the Taguchi method still does not provide optimal conditions.

The response surface methodology (RSM), can be used to solve the weaknesses of this traditional approach. By using a set of experimental trials, all parameters are varied simultaneously. Khayet et al. (2012) mentioned that, by applying RSM the number of the experimental trials can be minimized even though many UF condition parameters have been investigated at the same time compared to the trial and error optimization technique. Therefore, RSM is better compared to the familiar conventional optimization method. Some of the benefits of using RSM are the experiment becomes faster and flexible with just a small number of experiments that reduce time and related costs.

Modelling methods with a direct analysis of experimental data is an excellent option to the techniques that use phenomenological hypotheses such as knowledge-based models. In particular, particle swarm optimization (PSO) has been introduced in various fields such as environmental studies. PSO is an effective predictive method in the modeling for the behavior of nonlinear dynamic systems such as UF processes. In general, PSO refers to a class of distributed algorithms that have properties similar to self-structured interactions between several simple agents.

Hence, in this study, the optimization of FI conditions of PES UF hollow fiber membranes is required in two stages: (i) modeling of FI parameter relationship during UF process, and (ii) determine the optimal condition of FI. The factors that affect the FI during UF conditions are pH and temperature of wastewater solution, transmembrane pressure, and time during UF process. Design of experiments (DOE) integrated with the PSO methodology are used for this study. The DOE including central composite design (CCD) and RSM are used to develop the regression model of the FI condition. The regression model is used as an objective function in PSO in order to maximize the FI performance. Then, a PSO algorithm is developed to determine the optimum process parameters and system configurations. The PSO will determine the optimum settings for the parameters of FI during UF process. Hence, this research is needed to cover the gap of previous researches.

1.3 Research Questions

This research is done to give explanation to the three main questions which are:

i. What are the parameters and factors that influence the PES UF process performance?

ii. Which parameters or factors affect fouling index performance during UF process?

iii. What are the optimal fouling index conditions of UF PES hollow fiber membranes?

1.4 Research Objectives

This research consists of three main objectives which are:

i. To identify the factors and parameters that influence the UF PES process performance.

ii. To determine the significant fouling parameters and their relationship using RSM method.

iii. To optimize fouling conditions and optimize fouling index of PES UF hollow fiber membrane using PSO method.

1.5 Scopes of the Research

To accomplish all the objectives of this research, there are scope been chosen in this research. Several key areas of this research have been identified for optimizing the performance of PES UF hollow fiber membrane fouling.

i. A main polymeric material which is PES was chosen as is used in dope formulation.

ii. Fouling index is used to determine membrane performance during UF process.

iii. Synthesized oily wastewater is used as a main medium for characterizing the separation performance.

iv. Response surface methodology (RSM) is used to construct a regression model for identifying the relationship between fouling index performance and UF conditions.

v. PSO is used to define the optimal fouling conditions.

vi. MATLAB software version (R2020a) is used to implement the PSO optimization process.

vii. Parameters to investigate are pH, temperature of sample water, time of filtration process, surface area of membrane and transmembrane pressure.

1.6 Research Significant

Since water is a main resource for whole life, development strategies and sustainable control in reducing membrane fouling during the water filtration process is a very important mission. Based on this research, using the PSO optimization method, it demonstrates the success of reducing fouling membranes during the water filtration. Fouling control strategies can increase membrane life, energy demand, increase the lifespan of the membrane modules and the membrane maintenance besides the operation costs for to the membrane cleaning can be reduces. In addition, when membrane fouling reduced, membrane water filtration becomes more effective and efficient. This also an economic benefits approach which are reducing cost and money saving while producing good quality products of UF PES hollow fiber membranes with the desire properties. Therefore, this research could help future research to reduce membrane fouling, thus improving and contributing more sustain to the filtering system.

The UF process is very helpful in removal oily wastewater especially produced by the industry based on the accessibility to sustain the water supply system. The UF process is an excellent membrane separation because of the concept of separation which is focus on molecular size hence required lower operating costs compared to traditional methods. This research uses PES UF hollow fiber membrane because of PES is the most suitable type of membrane to treat oily wastewater. The performance of the UF process is measured by determining the value of FI. The value of FI is calculated based on the flux evaluated during the process. The impact of this study is important because it can reduce the PES UF hollow fiber membranes fouling since it offers a prospect of higher productivity and selectivity. Indirectly, this research can help to reduce the fouling of membranes, thus contributing to a more sustainable filtration system.

The final results acquired from this research are the optimal FI values during the membrane UF process by using the PSO methodology to reduce membrane fouling. This study can help engineers or decision makers to determine the appropriate way to solve the problem during the water filtration process in a short period of time. PSO helps to enhance the higher water flow during separation process, environmentally friendly, and requires little investment and energy consumption.

1.7 Structure of Research

This research consists of 5 chapters. Chapter 1 is the introduction to this study. Review of the topic, problem background, problem statement, study objectives, scope of study discussed. This chapter discusses the membrane filtration process in general and the membrane contamination process. Chapter 2 contains a literature review on PSO techniques and membrane technology. In membrane technology it is focused on membrane contamination at the time of filtration process. Chapter 3 discusses the proposed method framework for this study. This chapter discussed in detail the steps in the experiment that have been implemented. Chapter 4 describes the process to develop the regression model based on RSM and statistical regression techniques for the Fouling Index (FI). This chapter also discussed the optimization process of the FI using PSO and evaluates all of the findings and validates the experiments. Meanwhile, Chapter 5 explained the general conclusions of this research and some suggestions for the future research.

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LIST OF PUBLICATIONS

H. Norafifah, A.S. Noor Adila, M.Y. Noordin, K.Y. Wong and S. Izman. (2015). Process Clarification of Ultrafiltration of Hollow Fiber Membrane Using Design of Experiments Method. *ARPN Journal of Engineering and Applied Sciences*. (Scopus Indexed)

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