## Faculty of Mechanical Engineering

# ELECTROSPUN NYLON 6 NANOFIBRE WATER FILTRATION MEDIA FOR WASTEWATER TREATMENT

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**Master of Science in Mechanical Engineering**

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# ELECTROSPUN NYLON 6 NANOFIBRE WATER FILTRATION MEDIA FOR WASTEWATER TREATMENT

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**A thesis submitted**
**in fulfillment of requirements for the degree of Master of Science**
**in Mechanical Engineering**

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# DECLARATION

I declare that this thesis entitled "Electrospun Nylon 6 Nanofibre Water Filtration Media for Wastewater Treatment" is the result of my own research except as cited in the references. The thesis has not been accepted for any degree and is not concurrently submitted in candidature of any other degree.

Signature :…………………………………………………...

Name :…………………………………………………...

Date :…………………………………………………...

## APPROVAL

I hereby declare that I have read this thesis and in my opinion this thesis is sufficient in terms of scope and quality for the award of Master of Science in Mechanical Engineering.

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| Signature | : | ........................................................................ |
| Supervisor Name | : | ........................................................................ |
| Date | : | ........................................................................ |

# DEDICATION

To my beloved mother and father

**ABSTRACT**

Uncontrolled discharge of wastewater effluent has been a major concern due to the negative impact to human health and the environment. The issue becomes even more serious if the location of discharge point is located upstream of the river flow. Recently, increasingly stringent regulation has caused filter manufacturers to explore new materials to improve their products. Water filtration media using ultrafine fibres have attracted much attention among researchers due to their promising potentials. In this study, a new nylon 6 electrospun nanofibre incorporated water filtration media prepared by direct electrospinning was studied. The type of filtration media produced using this technique was nanofiltration. Electrospinning technique has been chosen in this study because of its simplicity and low cost. Using this technique, the quality of electrospun nanofibres will highly depend on electrospinning parameters during fabrication process. Therefore, the first objective of this study was to determine the optimum electrospinning parameters, namely electrospinning distance, solution concentration, and applied voltage. The second objective was to evaluate and characterise the filtration media based on results from the first experiment. Triplicate samples were prepared at different deposition times of nanofibre electrospunned to the substrate; 20 s, 40 s, 60 s, 80 s, 100 s and 120 s, to investigate the relationship between the amount of nanofibre and filtration efficiency. The performance of the samples were evaluated based on the filtration test using a vacuum pump in accordance with *BS EN 872:2005 Water Quality – Determination of Suspended Solids. Method by Filtration Through Glass Fibre Filters*. The water sample was obtained from the new UTeM's wastewater treatment plant. Suspended solid and chemical oxygen demand (COD) contents were also measured using a colorimeteric method in accordance with *Method 8006: HACH Method of Determining Suspended Solid* and *Method 8000: HACH Method of Determining Chemical Oxygen Demand*. The characterisation of the samples was carried out using scanning electron microscopy (SEM) to study the morphological structure of fibres, Energy Dispersive X-Ray Analyzer (EDX) element composition on the filter, ImageJ image analysis to determine average fibre diameter and mercury porosimetry to study porosity of the filters. From the first experiment, the optimum electrospinning distance, solution concentration and applied voltage were found to be at 10 cm, 20 wt.% and 14 kV respectively, based on average fibre diameter, morphological structure of fibres, and deposition amount. The second experiment suggests that the inclusion of electrospun nanofibre had significantly increased the efficiency of the filters. Suspended solid removal has increased to 100% at 80 s deposition time whilst COD has increased to 65% at 120 s deposition time, as compared to control sample (no nanofibre deposition) with 39.3% and 28.8% of suspended solid and COD both at 120 s deposition time, respectively. The effectiveness of the filter was also found to be improved when the amount of nanofibres increased. The inclusion of nanofibre layer provides an effective filtration medium due to their high surface area, small pore sizes and good interconnectivity of the pores. Furthermore, the inclusion of the nanofibres has little effect on the porosity of the developed filters. The knowledge gained from this study could extend the use of electrospun nanofibres in filtration applications. The knowledge would also facilitate new improvements and alternative solutions in any applications that require high surface area and high porosity filtration system.

# *ABSTRAK*

*Pelepasan efluen air sisa yang tidak terkawal telah menjadi kebimbangan utama kerana kesan negatif terhadap kesihatan manusia dan alam sekitar. Isu ini menjadi lebih serius jika lokasi titik pelepasan terletak di hulu aliran sungai. Baru-baru ini, peraturan yang semakin ketat telah menyebabkan pengeluar penapis untuk meneroka bahan baru untuk memperbaiki produk mereka. Media penapisan air yang menggunakan serat ultra halus telah menarik banyak perhatian di kalangan penyelidik kerana potensinya yang menjanjikan. Dalam kajian ini, sebuah media filtrasi air nilon 6 yang baru yang disediakan melalui teknik pemintalan elektro secara terus telah dikaji. Teknik ini menghasilkan media filtrasi jenis filtrasi nano. Teknik pemintalan elektro telah dipilih dalam kajian ini kerana kemudahan dan kosnya yang rendah. Menggunakan teknik ini, kualiti nanoserat elektrospun akan sangat bergantung kepada parameter elektrospinning semasa proses fabrikasi. Oleh itu, objektif pertama kajian ini adalah untuk menentukan parameter pemintalan elektro yang optimum, iaitu jarak pemintalan, kepekatan larutan, dan voltan yang digunakan. Objektif kedua adalah untuk menilai dan mencirikan media penapisan berdasarkan keputusan dari eksperimen pertama. Tiga sampel disediakan dengan memintal elektro nanoserat pada substrat pada masa pemendapan yang berbeza iaitu 20 s, 40 s, 60 s, 80 s, 100 s dan 120 s, untuk mengkaji hubungan antara jumlah nanoserat dan kecekapan penapisan. Kehandalan sampel dinilai menggunakan sistem penapisan pam vakum mengikut BS EN 872: 2005 Kualiti Air - Penentuan Jumlah Pepejal Terampai. Kaedah oleh Penapisan Melalui Penapis Serat Kaca. Sampel air diperoleh dari loji rawatan air sisa UTeM yang baru. Kandungan pepejal terampai dan permintaan oksigen kimia (COD) juga diukur dengan menggunakan kaedah colorimeterik mengikut Kaedah 8006: Kaedah Penentuan Pepejal Terampai HACH dan Kaedah 8000: Kaedah Penentuan Permintaan Oksigen Kimia HACH. Pencirian sampel dilakukan dengan menggunakan mikroskop pengimbasan elektron (SEM) untuk mengkaji struktur morphologi serat, Penganalisis Tenaga Serakan X-Ray Analyzer (EDX) untuk mengkaji komposisi elemen pada penapis, analisis imej ImageJ untuk menentukan purata diameter serat dan porosimetri merkuri untuk mengkaji keliangan pada penapis. Dari eksperimen pertama, jarak pemintalan yang optimum, kepekatan larutan dan voltan yang digunakan didapati pada 10 cm, 20% berat dan 14 kV masing-masing, berdasarkan purata diameter, struktur morfologi serat, dan jumlah pemendapan. Eksperimen kedua menunjukkan bahawa penambahan nanoserat elektrospun telah meningkatkan kecekapan penapis dengan ketara. Penyingkiran pepejal terampai telah meningkat kepada 100% pada 80 s masa mendapan manakala COD telah meningkat kepada 65% pada 120 s masa mendapan, berbanding dengan sampel kawalan (tiada nanoserat) dengan 39.3% dan 28.8% keduanya pada 120 s masa mendapan, masing-masing. Keberkesanan penapis juga didapati bertambah baik apabila jumlah nanoserat meningkat. Penambahan lapisan nanoserat menyediakan medium penapisan berkesan kerana luas kawasan permukaannya yang tinggi, saiz liang yang kecil dan hubungan yang baik dari liang ke liang. Selain itu, penambahan nanoserat tidak mempunyai kesan ke atas liang penapis. Maklumat diperoleh daripada kajian ini dapat memperluaskan penggunaan nanoserat elektrospun dalam aplikasi penapisan. Maklumat ini juga akan memudahkan penambahbaikan baru dan menjadi penyelesaian alternatif dalam mana-mana aplikasi yang memerlukan kawasan permukaan yang tinggi dan sistem penapisan keliangan yang tinggi.*

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## LIST OF ABBREVIATIONS

| | | |
|---|---|---|
| BOD | - | Biochemical oxygen demand |
| CNT | - | Carbon Nanotube |
| COD | - | Chemical oxygen demand |
| DMF | - | Dimethylformamide |
| DO | - | Dissolved oxygen |
| DWCNT | - | Double-wall Carbon Nanotube |
| FE-SEM | - | Filed Emission Electron Microscpe |
| HIPS | - | High-impact polystyrene |
| MWCNT | - | Multi-wall Carbon Nanotube |
| PAN | - | Polyacrylonitrile |
| PANi | - | Polyaniline |
| PCL | - | Poly(ε-caprolactone) |
| PEMFCs | - | Polymer electrolyte membrane fuel cells |
| PET | - | Polyethylene terephthalate |
| PGA | - | Poly(glycolic acid) |
| PLA | - | Poly(lactic acid) |
| PU | - | Polyurethanes |
| PVA | - | Polyvinyl Alcohol |
| PVDF | - | Polyvinylidene Flouride |
| SEM | - | Scanning Electron Microscope |
| SS | - | Suspended solid |
| STP | - | Sewage treatment plant |

TEM - Transmission Electron Microscopy

TSS - Total suspended solid

# LIST OF PUBLICATIONS

The research papers produced and published during the course of this research are as follows:

Journal:

1. N.S.A. Roslan, A. H. Nurfaizey, M.I. Mohamed Hafiz, M. Noryani, M. R. Mansor and N. A. Munajat, 2018. Nylon electrospun nanofibre water filtration media for wastewater treatment. *Materials Research Express – IOP Science*. *(Scopus).* (published)
2. N.S.A. Roslan, A.H. Nurfaizey, M.I. Mohamed Hafiz and R.. Nadlene, 2018. The Effect of Deposition Time on Filtration Efficiency of Electrospun Nanofibre Water Filters. *Journal of Advanced Manufacturing Technology (JAMT)*. (accepted)

Conference papers:

1. N.S.A. Roslan, A. H. Nurfaizey, M.I. Mohamed Hafiz, 2017. The Effect of Deposition Time on Filtration Efficiency of Electrospun Nanofibre Water Filters. *(ICE-SEAM).* (accepted)
2. N.S.A. Roslan, A.H. Nurfaizey, M.I. Mohamed Hafiz, M.R. Mansor, N.A. Munajat 2017. Fabrication and evaluation of nylon 6 electrospun nanofibre water filtration media for removing suspended solid. *Proceedings of Mechanical Engineering Research Day 2017*, Melaka, Malaysia, pp. 387-388. (published)

# CHAPTER 1

# INTRODUCTION

## 1.1 Research background

Recently, research and development on nanofibrous materials using electrospinning technique has been given a great attention by scientific and industrial community (Ramakrishna et al., 2005). Electrospinning technique is preferred over other nanofibre production techniques because of its simplicity, versatility, cost efficient and high potential applications. The process starts by charging a pendant droplet of polymer solution or melt with a high electric potential. When the electrostatic forces overcome the surface tension of the droplet at the tip of spinneret, a jet of fibre is erupted towards a grounded collector electrode. Initially the jet travels in a straight trajectory before transforming into a chaotic motion known as the whipping instability (Nurfaizey et al., 2014). Due to the whipping instability, random oriented fibres are collected at the collector electrode.

Nanofibres can be produced through several processing methods such as drawing, template synthesis, phase separation, self-assembly and electrospinning (Shao et al., 2012). In comparison with other nanofibre processing techniques, electrospinning is the only method that could potentially be developed for mass production (Ramakrishna et al., 2005). Electrospun nanofibres have been proposed for various applications such as in healthcare, biotechnology, environmental engineering, energy generation, protective clothing and electronics (Ramakrishna et al., 2006). In environmental engineering sector, electrospun nanofibers have been proposed for the development of high efficient water and air filtration systems (Balamurugan et al., 2011). Electrospun nanofibres are advantageous for filtration applications because of their good interconnectivity of pores and can be formed into highly porous mesh with high specific surface area (Ramakrishna et al., 2006). In fact, the use of nanofibrous membrane separation technology has been proven effective for removal of dyes, heavy metals and other contaminants in wastewater treatment process (Jie et al., 2015).

Rapid industrialization and urbanization have brought serious challenges to maintain good water quality especially in developing countries (Capps et al., 2015). Our country, Malaysia is not exempted from the arising water issues despite located in the tropical zone and receives high amount of rainfall (Johnson, 2016). The Malaysian Water Association reported that Malaysia is facing several water issues including inadequate water supply and degraded water quality at main water bodies such as rivers and lakes (Abdullah and Daud, 2013). Factories, power plants, and sewage treatment plants are the direct sources of water pollution since these premises release their waste to lakes or rivers (Islam et al., 2010). Various organic and inorganic impurities from these sources could contaminate the freshwater resources to a worrying level (Schwarzenbach et al., 2016). Chemical treatment is both economically advantageous and efficient. However, it requires an oxidation step if the metals are complexed (Crini et. al., 2019). Biological wastewater treatment facilities have been used to solve the issue, but with considerable success due to the presence of non-biodegradable and toxic contaminants (Grigory and Semiat, 2008). Physical filtration processes such as sequencing batch reactor could reduce the contaminants by transforming the contaminants from one phase to another but the process would create a highly concentrated sludge, which could be noxious and difficult to degrade (Bali et al., 2003; Anjum et al., 2016).

Researchers are continuously looking for a new way to improve water filtration process as an initiative to deal with the arising water issues. The use of new class of materials such as ultrafine nanofibres has gained much interest in filtration technology (Mokhena et al., 2015). Having plentiful benefits, electrospun nanofibres are being considered to be used in water filtration system to improve the efficiency of an existing filtration technology. Recent studies have shown that an advanced wastewater treatment system featuring electrospun nanofibres could be an effective solution to overcome the water issues (Kenry and Lim, 2017). A significant amount of effort has been dedicated by researchers in studying nanofibre water filters to remove various kind of contaminants. In a study by Xu et al. (2008), polysulfone electrospun fibre membrane was successfully used to reduce chemical oxygen demand (COD), ammonia and suspended solid (SS) from bio-treated wastewater. In another study, it was claimed that polyvinylidene fluoride electrospun nanofibre membrane could filtered about 90% of wastewater micro-particles (Kootenaei and Rad, 2013). Similar study from Thakura et al. (2015) also reported that more than 90% removal of COD from wastewater was achieved by using electrospun nanofibre membranes. Recently, Salahi et al. (2015) reported that polyethersulfone electrospun nanofibre membranes could effectively remove 83.1% of COD and 100% of SS. Some of these studies are reviewed in more detail in the next chapter.

This study focuses on fabrication, characterisation, and evaluation of new nylon 6 nanofibre incorporated water filters. Nylon 6 nanofibres were prepared using a laboratory scale electrospinning machine. The morphology of the filters was examined using scanning electron microscopy. The filters were characterised using elemental analysis and mercury porosimetry. The filters were evaluated based on the ability of the filter to reduce suspended solid and chemical oxygen demand. The water sample was taken from a sewage treatment plant at the Universiti Teknikal Malaysia Melaka, Durian Tunggal, Melaka which is located upstream of a highly populated area of Melaka City. The main hypothesis of this study is that the inclusion of nylon electrospun nanofibres onto commercial standard filter would significantly increase the filtration efficiency of the filter. The result gained from this study is important to provide a useful insight for improving the existing water filtration process.

### 1.2 Problem statement

A good water sources management is important to provide continuous clean water supply. One of the important measures is to improve the efficiency of wastewater treatment processes. Most treatment plants are built to clean wastewater before discharging the water into the water streams, or for reuse. Therefore, it is important to maintain the efficiency of the process to avoid toxics and dangerous contaminants from polluting the water stream. At the Universiti Teknikal Malaysia Melaka (UTeM), a new sewage treatment plant (STP) was built to treat water sewage and maintain the release of effluent in accordance to the regulation. According to the Eleventh Schedule of Akta Kualiti Alam Sekeliling 1974 (Kumbahan) 2009, a sewage treatment plant that is located at the upstream area of discharge point must comply with the Standard A of effluent discharge limits. On the other hand, Standard B will be applied for plants that discharge water at downstream of water intake area. In this case, the STP in UTeM should comply to the Standard A as it is located at the upstream of the water intake point.

Furthermore, the amount of wastewater that need to be treated is increasing due to rapid urbanization and industrialization. Untreated wastewater or improperly treated wastewater are harmful to be discharged to the water bodies. The sources of influent or water that need to be treated by the STP at UTeM are coming from 16 buildings in the main campus area. After a series of treatment, the effluent or treated wastewater will then be released to the Anak Sungai Garing located nearby the campus. At UTeM STP, five effluent quality parameters are consistently monitored namely COD, BOD, suspended solid, ammonia and pH value. However, among these parameters, the value of COD and suspended solid