

DEVELOPMENT AND CHARACTERIZATION OF POLYSULFONE - CARBON  
MOLECULAR SIEVES MIXED MATRIX MEMBRANE FOR  
OXYGEN / NITROGEN GAS SEPARATION

WAN RAFIZAH BINTI WAN ABDULLAH

UNIVERSITI TEKNOLOGI MALAYSIA



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## BORANG PENGESAHAN STATUS TESIS ♦

JUDUL : **DEVELOPMENT AND CHARACTERIZATION OF  
POLYSULFONE-CARBON MOLECULAR SIEVES MIXED  
MATRIX MEMBRANE FOR OXYGEN/NITROGEN GAS  
SEPARATION**

SESI PENGAJIAN: 2007/2008-I

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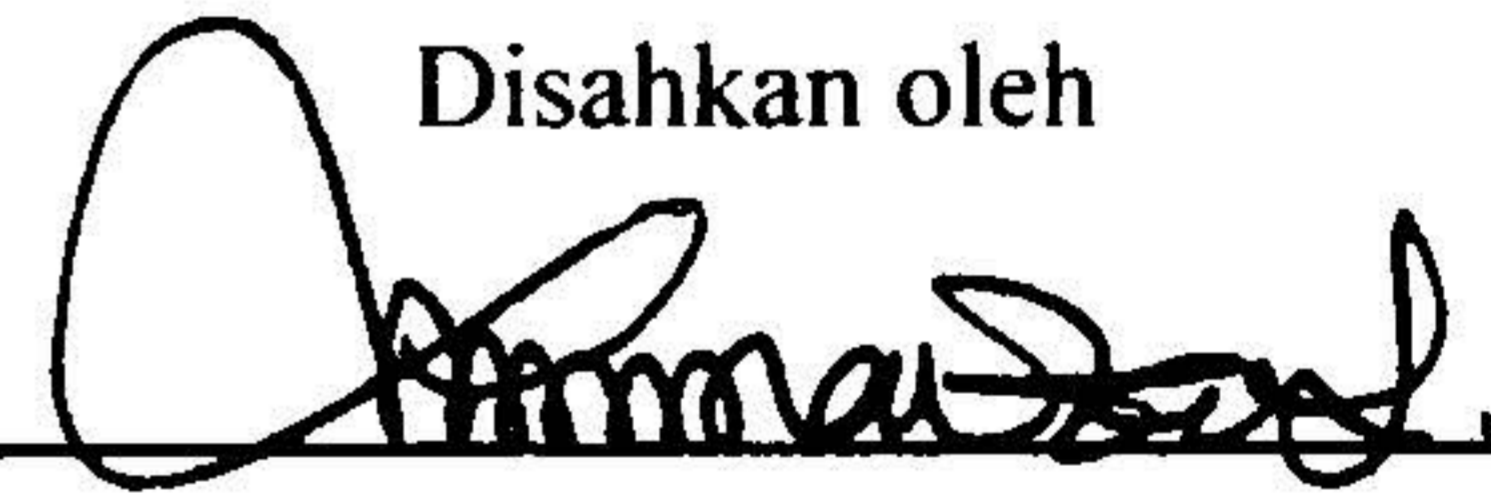
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Nama Penyelia

Tarikh: 10 SEPTEMBER 2007

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Date : 10<sup>th</sup> September 2007

## BAHAGIAN A – Pengesahan Kerjasama\*

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## BAHAGIAN B – Untuk Kegunaan Pejabat Sekolah Pengajian Siswazah (SPS)

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Nama : **GANESAN A/L ANDIMUTHU**

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PUSAT PEMBELAJARAN DIGITAL SULTANAH NUR ZAHIRAH

A thesis submitted in fulfilment of the  
requirements for the award of the degree  
Master of Engineering (Gas)

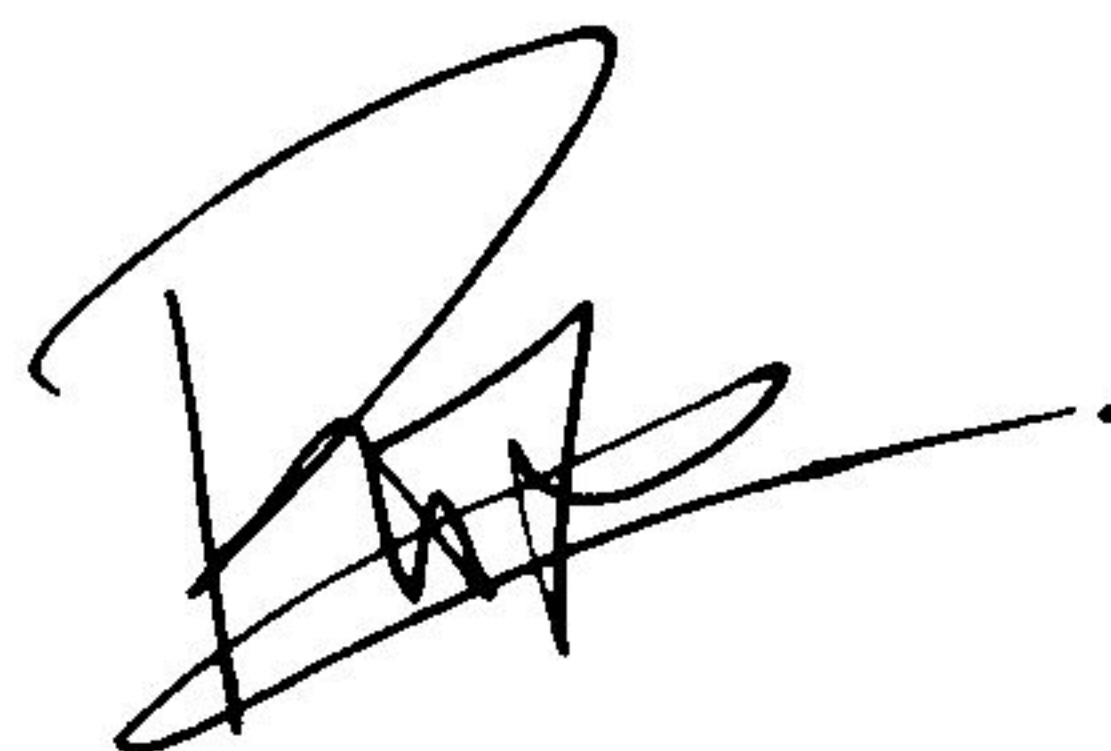
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SEPTEMBER 2007

I declare that this thesis entitled "*Development and Characterization of Polysulfone Carbon Molecular Sieve Mixed Matrix Membrane for Oxygen/Nitrogen Gas Separation*" is the result of my own research except as cited in references. The thesis has not been accepted for any degree and it is not concurrently submitted in candidature of any other degree.

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Name of Author

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WAN RAFIZAH BINTI WAN ABDULLAH

Date

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10<sup>th</sup> SEPTEMBER 2007

*Dedicated to my beloved family and friends who gave me inspiration, encouragement and endless support throughout the success of my study. May this thesis be an inspiration and guidance in the future.*



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## ABSTRACT

The objective of this study is to develop and characterize mixed matrix membranes (MMMs) containing carbon molecular sieves (CMS) in polysulfone (PSF), Udel<sup>®</sup> P-1700 matrix for oxygen/nitrogen gas separation. The MMMs were fabricated in the form of dense film by incorporating CMS particles (< 25  $\mu\text{m}$ ) into 20 wt% PSF in n-methylpyrrolidone (NMP) using solution casting method. The membrane characterization was performed using Field Emission Scanning Electron Microscopy (FESEM), Differential Scanning Calorimetry (DSC), Thermal Gravimetry Analysis (TGA), Attenuated Total Reflection Fourier Transform Infrared (ATR-FTIR) methods and single gas permeation test. Effects of CMS loadings on the properties of MMMs were studied by varying the concentration of CMS from 10 to 35 wt%. Effects of CMS sizing with poly (vinyl pyrrolidone) (PVP) K-15 prior to embedment into the polymer matrix on the properties of MMMs were also investigated. The oxygen permeability and nitrogen permeability increased with increasing CMS loadings. The oxygen/nitrogen selectivity increased up to 20 wt% loading of CMS and then decreased as the CMS loadings were further increased. The interfacial contact between CMS and PSF matrix was excellent at low CMS loadings (10 - 20 wt%). However, the presence of 'sieve-in-a-cage' morphology became more prominent at higher CMS loadings (30 - 35 wt%) causing a considerable reduction in the separation performance. The highest oxygen/nitrogen selectivity achieved was 5.97, obtained with 20 wt% CMS loading at feed pressure of 1.5 bar. The sizing of CMS using PVP K-15 improved the interfacial adhesion between CMS and PSF matrix. As a result, the PSF-PVP K-15 sized CMS MMMs exhibited oxygen/nitrogen selectivity of 6.05 compared to that of the unmodified MMM which was about 3.69. This study proposed that a proper fabrication method and an appropriate combination of polymer and sieving media could ensure the successful formation of mixed matrix membrane with the absence of defects and exhibit attractive separation properties.

## ABSTRAK

Objektif kajian ini adalah untuk membangunkan dan mencirikan membran matrik tercampur (MMMs) yang mengandungi karbon penapis molekul (CMS) di dalam matrik polisulfona (PSF), Udel<sup>®</sup> P-1700 bagi pemisahan oksigen/nitrogen. MMMs dihasilkan dalam bentuk filem terpadat dengan mencampurkan patikel CMS (< 25  $\mu\text{m}$ ) ke dalam larutan 20 % jisim PSF dalam n-metilpirolidon (NMP) melalui kaedah tuangan larutan. Pencirian membran dijalankan menggunakan kaedah Kemikroskopan Elektron Imbasan-Pemancaran Lapangan (FESEM), Kalorimetri Pembezaan Imbasan (DSC), Analisis Gravimetri Terma (TGA), spektroskopi Inframerah Penjelmaan Fourier (ATR-FTIR) dan ujian ketelapan gas tunggal. Kesan kandungan CMS terhadap ciri-ciri MMMs dikaji dengan merubah kepekatan CMS dari 10 ke 35 % jisim. Kesan pemerekatan CMS menggunakan polivinilpirolidon (PVP) K-15 sebelum dicampurkan ke dalam matrik polimer terhadap ciri-ciri MMMs juga telah dikaji. Kebolehtelapan oksigen dan nitrogen meningkat dengan peningkatan kandungan CMS. Kememilihan oksigen/nitrogen meningkat sehingga ke 20 % jisim CMS dan kemudiannya menurun apabila kandungan CMS terus ditingkatkan. Pelekatan antaramuka CMS dan matrik PSF adalah sangat baik pada kandungan CMS yang rendah (10 - 20 % jisim). Namun, kewujudan morfologi '*penapis di dalam kurungan*' menjadi semakin jelas pada kandungan CMS yang tinggi (30 – 35 % jisim) menyebabkan pengurangan ketara dalam prestasi pemisahan. Kememilihan tertinggi yang dicapai adalah 5.97, diperolehi dengan kandungan CMS 20 % jisim pada tekanan masukan 1.5 bar. Pemerekatan CMS menggunakan PVP K-15 mempertingkatkan pelekatan antara muka CMS dan matrik PSF. Kesannya, MMM PSF-CMS terekat PVP K-15 menunjukkan kememilihan oksigen/nitrogen sehingga 6.05 berbanding MMM takterubahsuai iaitu lebih kurang 3.69. Kajian ini mencadangkan bahawa kaedah fabrikasi yang sesuai dan kombinasi polimer dan bahan penapis yang tepat akan menghasilkan membran matrik tercampur tanpa kecacatan dengan ciri-ciri pemisahan yang menarik.