



**UNIVERSITI PUTRA MALAYSIA**

***PURIFICATION OF GLYCOLYTIC PRODUCT FROM POLYETHYLENE  
TEREPHTHALATE (PET) WASTE BY A TWO-STAGE EVAPORATION  
PROCESS***

**GOH HUI WEN**

**FK 2012 8**

**PURIFICATION OF GLYCOLYTIC PRODUCT FROM POLYETHYLENE  
TEREPHTHALATE (PET) WASTE BY A TWO-STAGE EVAPORATION  
PROCESS**



**By**

**GOH HUI WEN**

**Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in  
Fulfillment of the Requirements for the Degree of Master of Science**

**APRIL 2012**

Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfillment of the requirement for the degree of Master of Science.

**PURIFICATION OF GLYCOLYTIC PRODUCT FROM POLYETHYLENE TEREPHTHALATE (PET) WASTE BY A TWO-STAGE EVAPORATION PROCESS**

By

**GOH HUI WEN**

**April 2012**

**Chairman: Assoc. Prof. Salmiaton Ali, PhD**

**Faculty: Faculty of Engineering**

Malaysia is a developing country. With the rise in income and standard of living together with the rate of industrial growth in Malaysia, the demand of materials will keep increasing. This directly increases the amount of waste generated. In Malaysia, almost all type of solid wastes are disposed off into landfill sites. Due to the limitation of the available landfill sites, many solutions are introduced by the Malaysia government to reduce the solid wastes being dumped to the landfill. Polyethylene terephthalate (PET) bottle is one of the plastic solid wastes that can be easily found in Malaysia. One of the solutions that can be used to solve the abundant of PET wastes is chemical recycling of PET wastes to produce other value added product. This method not only can decrease the PET waste in landfill sites but also can produce many useful recycled PET products.

This research is focusing on the purification processes used in chemical recycling of PET waste. Crystallization and two stages evaporation processes were selected to purify the contaminated bis(2-hydroxyethyl) terephthalate (BHET) obtained from glycolysis of PET waste. This research was divided into two parts: first part was simulation of the two stages evaporation and crystallization processes using ASPEN PLUS to investigate the

effect of operating temperature and pressure of two stages evaporation toward the percentage of ethylene glycol (EG) removed, heat duty needed and percentage of BHET recovered. Based on the simulation findings, the first stage evaporation was simulated at pressure range of 130 Pa to 10,000 Pa and temperature range of 90 °C to 180 °C while the second stage evaporation was simulated at pressure range of 50 Pa to 250 Pa and temperature range of 120 °C to 180 °C. The crystallization was simulated at temperature range of 5 °C to 30 °C to study the effect of crystallization temperature toward the percentage of BHET recovered. The second part of the research was verification of simulation result by conducting experiments using conventional crystallization and two stages evaporation processes.

The ASPEN PLUS simulation results showed that increasing the operating temperature and decreasing the operating pressure of the two stages evaporation might increase the percentage of EG removed and at the same time increased the heat duty required and reduced the percentage of BHET recovered. The optimum conditions was selected based on higher EG removed with lower heat duty needed and higher BHET recovered. Optimum conditions of first and second stage evaporation were 105 °C and 1000 Pa and 130 °C and 50 Pa respectively. Two stages evaporation process was capable to reduce the composition of EG in glycolized mixture from 77 % to 0.15 % while increase the composition of BHET from 19 % to 82.48 %. Crystallization process using second purification route at 1 °C, with ratio of water used to glycolized solid of 5:1 and 3 hours cooling time was capable to remove EG yielding white crystallized solid consists of 93.02 % BHET. This shows that both methods can be used to purify glycolysis product.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Master Sains.

**PENULENAN PRODUK GLIKOLISIS DARIPADA SISA POLIETILENA  
TEREPHTHALATE (PET) OLEH PROSES DUA-PERINGKAT PENYEJATAN**

Oleh

**GOH HUI WEN**

**April 2012**

**Pengerusi: Prof. Madya Salmiaton Ali, PhD**

**Fakulti: Fakulti Kejuruteraan**

Malaysia merupakan sebuah negara yang sedang membangun. Kenaikan pendapatan dan taraf hidup rakyat Malaysia termasuk kenaikan kadar pertumbuhan industri menyebabkan permintaan terhadap bahan mentah seperti petrol, makanan, plastik dan lain-lain lagi meningkat. Ini akan menyebabkan jumlah penghasilan sampah meningkat. Di Malaysia, hampir semua sampah berbentuk pepejal dibuangkan ke tapak pelupusan. Oleh sebab tapak pelupusan terhad, dengan itu kerajaan Malaysia telah melaksanakan banyak kaedah untuk menyelesaikan masalah pertambahan sisa sampah berbentuk pepejal. Polyethylene terephthalate (PET) botol merupakan salah satu jenis plastik sisa pepejal yang senang dijumpai di merata tempat. Salah satu cara penyelesaian yang boleh digunakan untuk menyelesaikan masalah PET sisa bahan buangan ialah kitar semula PET sisa buangan secara kimia untuk menghasilkan bahan yang berguna. Cara ini bukan sahaja boleh mengurangkan PET sisa buangan di tapak pelupusan tetapi juga boleh menghasilkan banyak jenis bahan PET yang berguna.

Penyelidikan ini menumpukan pada kaedah penulenan yang digunakan dalam kaedah kitar semula sisa PET buangan secara kimia. Kaedah penghabluran secara konvensional

dan peyejatan proses secara dua peringkat telah digunakan untuk menuliskan bis(2-hydroxyethyl) terephthalate (BHET) yang diperolehi dari PET sisa buangan glikolisis proses.

Penyelidikan ini telah dibahagikan kepada dua bahagian: bahagian pertama ialah simulasi peyejatan dan penghabluran proses dengan menggunakan ASPEN PLUS untuk mengkajikan hubungan antara operasi suhu dan tekanan dan peratusan penyingkiran ethylene glycol (EG), duti haba yang diperlukan dan peratusan pemulihan BHET. Simulasi data menunjukkan peringkat pertama peyejatan proses boleh beroperasi pada 130 Pa ke 10,000 Pa dan 90 °C ke 180 °C manakala peringkat kedua peyejatan proses boleh beroperasi pada 50 Pa ke 250 Pa dan 120 °C ke 180 °C. Penghabluran proses dijalankan pada suhu 5 °C ke 30 °C untuk mengkajikan perubahan suhu penghabluran terhadap peratusan pemulihan BHET. Bahagian kedua merupakan pengesahan simulasi data dengan menjalankan eksperimen.

ASPEN PLUS simulasi data menunjukkan peratusan penyingkiran EG dan duti haba yang diperlukan menambah manakala peratusan pemulihan BHET menurun apabila operasi suhu meningkat dan operasi tekanan menurun bagi peyejatan proses. Keadaan optimum bagi peyejatan proses dipilih dengan merujuk pada maksimum peratusan penyingkiran EG dan minimum duti haba serta maksimum peratusan pemulihan BHET.

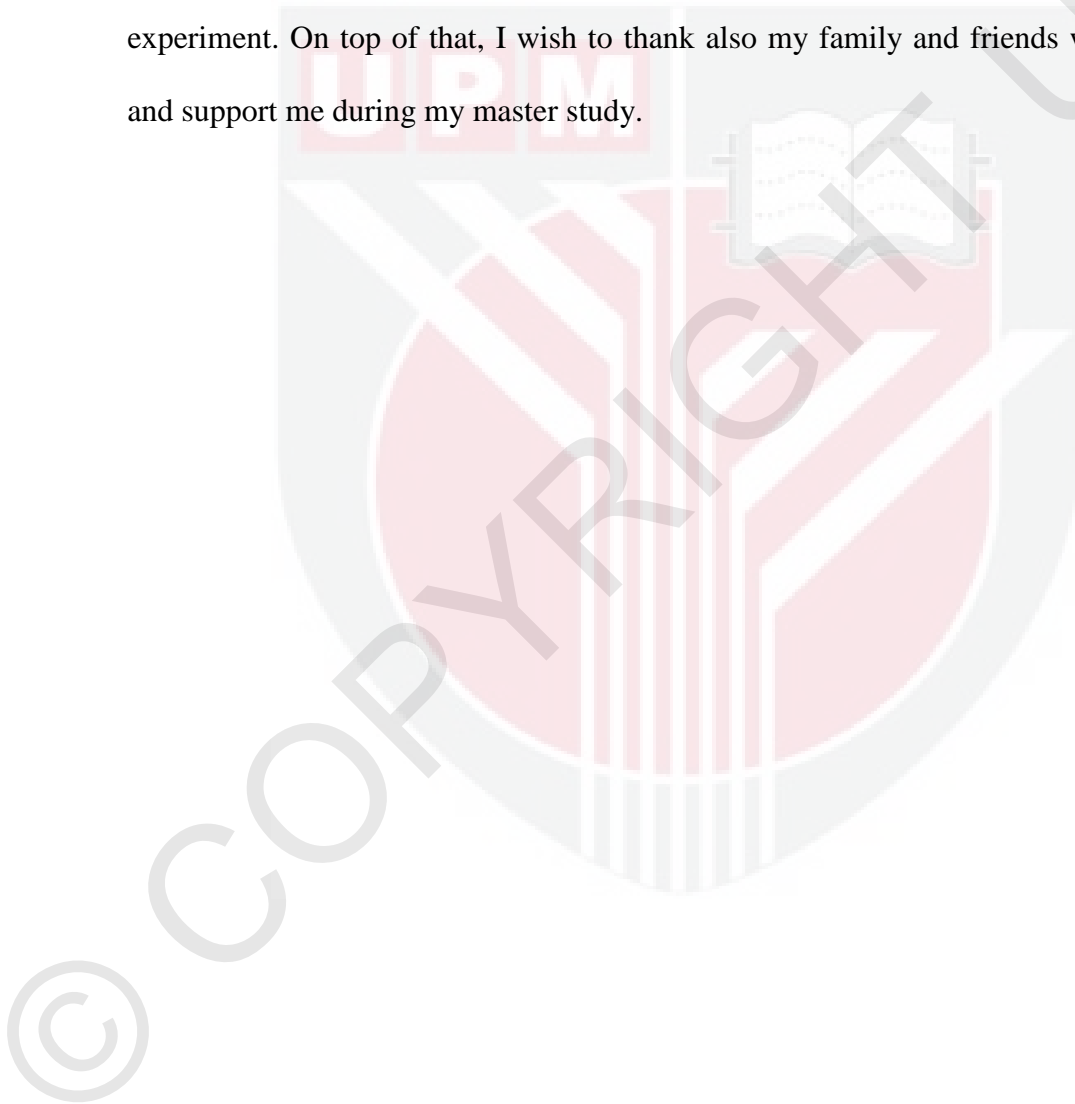
Keadaan optimum bagi peringkat pertama dan kedua proses peyejatan ialah 105 °C dan 1000 Pa serta 130 °C dan 50 Pa masing-masing. Dua peringkat proses peyejatan mampu mengurangkan komposisi EG dalam campuran *glycolyzed* daripada 77% kepada 0.15% manakala meningkatkan komposisi BHET daripada 19% kepada 82.48%. Proses penghabluran menggunakan laluan penulenan kedua pada 1 °C, dengan nisbah air yang

digunakan kepada pepejal glycolyzed 5:1 dan 3 jam masa penyejukan mampu membuang EG untuk menghasilkan pepejal kristal putih mengandungi 93.02%. Ini menunjukkan bahawa kedua-dua cara boleh digunakan untuk menuliskan produk glikolisis.



## ACKNOWLEDGEMENTS

I wish to thank Associate Professor Dr. Salmiaton bt. Ali for her significant contribution to this research and Associate Professor Dr. Norhafizah bt. Hj. Abdullah and Professor Dr. Azni b. Idris for their valuable advice in making this research successful. Besides that, I would like to thank also lab technicians for their assistance during carrying experiment. On top of that, I wish to thank also my family and friends who encourage and support me during my master study.





I certify that a Thesis Examination Committee has met on 23 April 2012 to conduct the final examination of GOH HUI WEN on her thesis entitled "**Purification of Glycolytic Product from Polyethylene Terephthalate Waste by a Two-stage Evaporation Process**" in accordance with the Universities and University Colleges Act 1971 and the Constitution of the Universiti Putra Malaysia [P.U.(A) 106] 15 March 1998. The Committee recommends that the student be awarded the Master of Science.

Members of the Thesis Examination Committee were as follows:

**Dayang Radiah bt Awang Biak, PhD**

Faculty of Engineering  
Universiti Putra Malaysia  
(Chairman)

**Thomas Choong Shean Yaw, PhD**

Professor and Ir  
Faculty of Engineering  
Universiti Putra Malaysia  
(Internal Examiner)

**Mohd. Halim Shah Ismail, PhD**

Associate Professor and Head of Department of Chemical and Environmental Engineering  
Faculty of Engineering  
Universiti Putra Malaysia  
(Internal Examiner)

**Ishak Bin Ahmad, PhD**

Associate Professor and Head of Chemical Programme  
Faculty of Science and Technology  
Universiti Kebangsaan Malaysia  
Malaysia  
(External Examiner)

---

**BUJANG KIM HUAT, PhD**

Professor and Dean  
School of Graduate Studies  
Universiti Putra Malaysia

Date:

This thesis was submitted to the Senate of Universiti Putra Malaysia and has been accepted as fulfillment of the requirement for the degree of Master of Science, Process Engineering. The members of the Supervisory Committee were as follows:

**Salmiaton bt. Ali, PhD**

Associate Professor  
Faculty of Engineering  
Universiti Putra Malaysia  
(Chairman)

**Norhafizah bt. Hj. Abdullah, PhD**

Associate Professor  
Faculty of Engineering  
Universiti Putra Malaysia  
(Member)

**Azni b. Idris, PhD**

Professor  
Faculty of Engineering  
Universiti Putra Malaysia  
(Member)

---

**BUJANG BIN KIM HUAT, PhD**

Professor and Dean  
School of Graduate Studies  
Universiti Putra Malaysia

Date:

## DECLARATION

I declare that the thesis is my original work except for quotations and citations which have been duly acknowledged. I also declare that it has not been previously, and is not concurrently, submitted for any other degree at Universiti Putra Malaysia or at any other institution.



---

**GOH HUI WEN**

Date: 23 April 2012



# TABLE OF CONTENT

	<b>Page</b>
<b>ABSTRACT</b>	ii
<b>ABSTRAK</b>	iv
<b>ACKNOWLEDGEMENTS</b>	vii
<b>APPROVAL</b>	viii
<b>DECLARATION</b>	x
<b>LIST OF TABLES</b>	xiv
<b>LIST OF FIGURES</b>	xviii
<b>LIST OF ABBREVIATIONS</b>	xxv
<b>CHAPTER</b>	
<b>1 INTRODUCTION</b>	
1.1 Research Background	1
1.2 Problem Statement	3
1.3 Objectives	4
1.4 Scope of Work	5
1.5 Thesis Outline	5
<b>2 LITERATURE REVIEW</b>	
2.1 Plastics Industry in Malaysia	6
2.2 Plastics Waste in Malaysia	8
2.3 Environmental Issues related to Plastic Wastes	9
2.4 PET and Its Monomer	10
2.5 PET Applications	13
2.6 Recycling of PET Wastes	15
2.6.1 Primary Recycling	16
2.6.2 Secondary Recycling	17
2.6.3 Tertiary Recycling	18
2.7 Impurities in PET Waste	24
2.8 BHET Purification	26
2.9 Application for Recycled PET/BHET	33
2.10 Summary of Chapter 2	34
<b>3 METHODOLOGY</b>	
3.1 Research Design	36
3.2 ASPEN PLUS Simulation	38
3.2.1 Single Stage Evaporation	38
3.2.1.1 Assumptions	38
3.2.1.2 Process Description	38
3.2.2 Two Stages Evaporation	43
3.2.2.1 Assumptions	44
3.2.2.2 Process Description	44
3.2.3 Crystallization using second purification route	45
3.2.3.1 Assumptions	46

3.2.3.2	Process Description	46
3.3	Response Surface Methodology (RSM)	48
3.4	Verification of Simulation Results	50
3.4.1	Materials	50
3.4.1.1	Raw Materials	50
3.4.1.2	Chemicals	50
3.4.1.3	Equipment	50
3.4.2	Glycolysis	51
3.4.3	Two Stages Evaporation	53
3.4.4	Crystallization	56
3.5	Process Routes	57
3.6	Product Analysis	60
3.6.1	DSC Analysis	60
3.6.2	HPLC Analysis	60
3.6.3	SEM Analysis	62
<b>4</b>	<b>RESULTS AND DISCUSSIONS</b>	
4.1	Introduction	63
4.2	Feedstock Composition	63
4.2.1	Glycolyzed Mixture and Glycolyzed Solid	64
4.2.2	GP Product	65
4.3	Simulation	68
4.3.1	Two Stages Evaporation	69
4.3.1.1	First Stage Evaporation	69
4.3.1.2	Second Stage Evaporation	74
4.3.2	Crystallization	80
4.3.3	Comparison between Two Stages Evaporation and Crystallization	81
4.3.4	Comparison between Single and Two Stages Evaporation	82
4.3.5	RSM for Two Stages Evaporation	90
4.3.5.1	RSM for First Stage Evaporation	90
4.3.5.2	RSM for Second Stage Evaporation	95
4.4	Verification of Simulated Results	99
4.4.1	Evaporation	100
4.4.1.1	First Stage Evaporation	100
4.4.1.2	Comparison between Simulation and Experimental Result	102
4.4.2	Crystallization	104
4.4.2.1	Crystallization using Second Purification Route	104
4.4.2.1.1	Crystallization Period	104
4.4.2.1.2	Crystallization Temperature	107
4.4.2.1.3	Ratio of Water Used to Glycolyzed Solid	109
4.4.2.2	Comparison between Simulation and Experimental Result	112
4.4.2.3	Crystallization using Third Purification Route	114
4.5	Summary of Simulation and Experimental Result	116

4.5.1	Summary of Simulation Result for Two Stages Evaporation	117
4.5.2	Summary of Experimental Result for Crystallization using Second Purification Route	119
<b>5</b>	<b>CONCLUSION AND RECOMMENDATIONS FOR FUTURE RESEARCH</b>	
5.1	Conclusion	120
5.2	Recommendations	122
	<b>REFERENCES</b>	123
	<b>APPENDIX A</b>	131
	<b>APPENDIX B</b>	132
	<b>APPENDIX C</b>	161
	<b>APPENDIX D</b>	168
	<b>APPENDIX E</b>	179
	<b>APPENDIX F</b>	210
	<b>BIODATA OF STUDENT</b>	211