UNIVERSITI PUTRA MALAYSIA

PREPARATION AND CHARACTERIZATION OF HYDROGELS FROM CARBOXYMETHYL CELLULOSE AND 1- VINYL-2-PYRROLIDONE USING IRRADIATION TECHNIQUES FOR SLOW RELEASE APPLICATION

NORHANIFAH BINTI MOHD YAZID

FS 2011 96
PREPARATION AND CHARACTERIZATION OF HYDROGELS FROM CARBOXYMETHYL CELLULOSE AND 1- VINYL-2-PYRROLIDONE USING IRRADIATION TECHNIQUES FOR SLOW RELEASE APPLICATION

NORHANIFAH BINTI MOHD YAZID

MASTER OF SCIENCE
UNIVERSITI PUTRA MALAYSIA
2011
PREPARATION AND CHARACTERIZATION OF HYDROGELS FROM CARBOXYMETHYL CELLULOSE AND 1- VINYL-2-PYRROLIDONE USING IRRADIATION TECHNIQUES FOR SLOW RELEASE APPLICATION

By

NORHANIFAH BINTI MOHD YAZID

Thesis Submitted to School of Graduate Studied, Universiti Putra Malaysia, in Fulfilments of the Requirements for the Degree of Master of Science.

February 2011
Abstract of thesis presented to the Senate of University Putra Malaysia in fulfilment of the requirement for the degree of the Master of Science

PREPARATION AND CHARACTERIZATION OF HYDROGELS FROM CARBOXYMETHYL CELLULOSE AND 1-VINYL-2-PYRROLIDONE USING IRRADIATION TECHNIQUES FOR SLOW RELEASE APPLICATION

By

NORHANIFAH BINTI MOHD YAZID

February 2011

Chairman : Associate Professor Mansor bin Ahmad, PhD
Faculty : Science

Hydrogels from carboxymethyl cellulose (CMC) and 1-vinyl-2-pyrrolidone (VP) were prepared via electron beam and ultraviolet radiations. Optimization of the preparation parameters was carried out for both methods where the optimum stirring time and percentage of crosslinking agents were at 3 hour of stirring and 5 % of BIS respectively. The hydrogels were prepared by irradiating CMC/VP samples with electron beam at 5, 10, 15, 20 and 25 kGy irradiation doses. For ultraviolet radiation, the optimum irradiation time and percentage of photoinitiator were at 8 hour of exposure and 1 % of photoinitiator. The effect of electron beam dose and concentrations of CMC on gel fraction, swelling behaviour, thermal properties and surface morphology were studied. When the irradiation dose and the concentration of CMC were increased, the gel fraction increased. Increasing the irradiation dose or the concentration of CMC resulted in the decrease of degree of swelling. The highest degree of swelling was obtained in alkaline medium followed by distilled water, salt
and acidic media. In the temperature-swelling studies, the highest degree of swelling was recorded at room temperature (25 °C). The FTIR analysis showed there was an intermolecular interaction between C=O and O-H which could be due to the intermolecular hydrogen bonding of carboxylic group and non-substituted hydroxyl groups in the CMC. Thermogravimetric analysis and differential thermogravimetric of hydrogels showed that thermal stability of hydrogels increased with increasing irradiation dose, but decreased when the concentration of CMC was increased. Differential scanning spectroscopy thermograms showed that melting temperature of hydrogels was affected by irradiation dose and concentration of CMC. The surface morphology study showed the pore size of the hydrogels was dependent on irradiation dose and concentration of CMC which affected the crosslinking density of the hydrogels. For the controlled release study, the highest released obtained from hydrogels irradiated at 10 kGy. For the ultraviolet radiation study, the gel fraction obtained was less than electron beam radiation and the degree swelling of hydrogels decreased with increasing gel fraction. The thermal properties showed that the melting temperatures of the hydrogels decreased compared to pure CMC and VP. The surface morphology study showed the pores size obtained after ultraviolet radiation was heterogeneous. Overall analysis revealed that electron beam radiation was the better technique compared to the ultraviolet radiation for preparing CMC/VP hydrogels.
PENYEDIAAN DAN PENCIRIAN HIDROGEL DARIPADA KARBOKSILMETIL SELULOSA DAN 1-VINIL-2-PYRROLIDONE MENGGUNAKAN TEKNIK-TEKNIK RADIASI UNTUK APLIKASI PENGALIRAN PERLAHAN

Oleh

NORHANIFAH BINTI MOHD YAZID

Februari 2011

Pengerusi : Profesor Madya Mansor bin Ahmad, PhD

Fakulti : Sains

ACKNOWLEDGEMENTS

First of all, thanks and Alhamdullillah to Allah for giving me faith and strength to complete my project.

I would like to express my deepest gratitude to Assoc. Prof. Dr. Mansor B. Hj. Ahmad for his supervisions and guidance throughout this study. He is rightfully top the list who has carried me through this study. Thank also to Dr. Kamaruddin Hashim and Dr. Norazowa Bt. Ibrahim. Without their guidances, advices and undying efforts, surely my project would not success.

I also would like to take this opportunity to express uncountable thanks to my parents; Mohd Yazid B. Noh and Fauziah Bt. Ismail, my siblings for their supports and encouragements with spare me the strength to finish this project.

Special thanks must go to all Chemistry Department’s staffs especially Mrs Zaidina and Mrs Norazlina, also to Agency Nuclear Malaysia’s staffs ; Miss Sabariah, Miss Nurain and Miss Nor Akmal who are always there with their technical expertise.

My friends and all polymer group members, thanks for your support, faith and always with me up and down. Thanks also Mr Muhammad Firdaus B. Haji Dasuki and I really appreciated the encouragement and understanding given by him during doing this study.
Last but not the least, thanks to Ministry of Science, Technology and Innovation (MOSTI) and Universiti Putra Malaysia (UPM) for facilities financial support during my study.
I certify that an Examination Committee met on date on viva to conduct the final examination of Norhanifah Bt. Mohd Yazid on her Master of Degree thesis entitle “Preparation and Characterization of Hydrogel from Carboxymethyl Cellulose and 1-Vinyl-2-Pyrrolodone using Irradiation Techniques for Slow Release Application” in accordance with Universiti Pertanian Malaysia (Higher Degree) Act 1980 and Universiti Pertanian Malaysia (Higher Degree) Regulations 1981. The Committee recommends that the student be awarded the relevant degree.

Members of the Examination Committee were as follows:

**ANNUAR KASSIM, Ph.D**  
Professor  
Faculty of Science  
Universiti Putra Malaysia  
(Chairman)

**MOHAMAD ZAKI AB.RAHMAN, Ph.D**  
Associate Professor  
Faculty of Science  
Universiti Putra Malaysia  
(Internal Examiner)

**SIDIK SILONG, Ph.D**  
Associate Professor  
Faculty of Science  
Universiti Putra Malaysia  
(Internal Examiner)

**RUSLI DAIK, Ph.D**  
Associate Professor  
School of Chemical Science and Food Technology  
Faculty of Science and Technology  
Universiti Kebangsaan Malaysia  
(External Examiner)

____________________________________  
BUJANG KIM HUAT, Ph.D  
Professor and Deputy Dean  
School of Graduate Studies  
Universiti Putra Malaysia

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

**Mansor Ahmad, PhD**  
Associate Professor  
Faculty of Science  
Universiti Putra Malaysia  
(Chairperson)

**Kamaruddin Hashim, PhD**  
Senior Researcher  
Agency Nuclear Malaysia  
Ministry of Science, Technology and Innovation, Malaysia  
(Member)

**Nor Azowa Ibrahim, PhD**  
Lecturer  
Faculty of Science  
Universiti Putra Malaysia  
(Member)

_______________________  
HASANAH MOHD GHAZALI, PhD  
Professor/Dean  
School of Graduate Studied.  
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 any other institution.

NORHANIFAH BINTI MOHD YAZID
Date: 9 February 2011
# ABLE OF CONTENTS

<table>
<thead>
<tr>
<th>Section</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>ABSTRACT</td>
<td>liii</td>
</tr>
<tr>
<td>ABSTRAK</td>
<td>v</td>
</tr>
<tr>
<td>ACKNOWLEDGEMENTS</td>
<td>vii</td>
</tr>
<tr>
<td>APPROVAL</td>
<td>ix</td>
</tr>
<tr>
<td>DECLARATION</td>
<td>xi</td>
</tr>
<tr>
<td>LIST OF TABLES</td>
<td>xiv</td>
</tr>
<tr>
<td>LIST OF FIGURES</td>
<td>xv</td>
</tr>
<tr>
<td>LIST OF SCHEMES</td>
<td>xix</td>
</tr>
<tr>
<td>LIST OF ABBREVIATIONS</td>
<td>xx</td>
</tr>
</tbody>
</table>

## CHAPTER

1. **INTRODUCTION**
   - 1.1 General background 1
   - 1.2 Background of the research 3
   - 1.3 Statements of the problem 5
   - 1.4 Objectives 7

2. **LITERATURE REVIEWS**
   - 2.1 Polymer 8
   - 2.2 Cellulose 9
     - 2.2.1 Carboxymethyl cellulose 10
   - 2.3 Poly (vinyl pyrrolidone) 12
   - 2.4 Hydrogels 13
   - 2.5 Application of the hydrogels 15
   - 2.6 Interpenetrating polymer networks 18
   - 2.7 Radiation of polymers 19
     - 2.7.1 Electron beam radiation 20
     - 2.7.2 Preparation hydrogels via electron beam radiation 22
     - 2.7.3 Ultraviolet radiation 23
     - 2.7.4 Preparation hydrogels via ultraviolet radiation 24
   - 2.8 General properties of hydrogels 26
     - 2.8.1 Degree of swelling 26
     - 2.8.2 Gel fraction 28
     - 2.8.3 Controlled release 30
     - 2.8.4 Thermal properties 31

3. **MATERIALS AND METHODS** 35
   - 3.1 Materials 35
   - 3.2 Methodology 35
     - 3.2.1 Optimization and preparation of hydrogels by electron beam radiation 36
     - 3.2.2 Optimization and preparation of hydrogels by ultraviolet radiation 38
   - 3.3 Properties Assessment 39
     - 3.3.1 Determination of gel fraction of hydrogels 3940

xii
3.3.2 Swelling behaviour of hydrogels

3.4 Characterization of the hydrogels
   3.4.1 Fourier Transform Infrared Analysis (FTIR)
   3.4.2 Thermogravimetric Analysis (TGA)
   3.4.3 Differential Scanning Calorimetry (DSC)
   3.4.4 Scanning Electron Microscopy (SEM)
   3.4.5 Controlled release test

4. RESULT AND DISCUSSION
   4.1 Preparation of optimization hydrogel by electron beam radiation
      4.1.1 The effect of stirring time on gel fraction
      4.1.2 The effect of crosslinking agent on gel fraction
      4.1.3 The effect of stirring time on swelling behaviour
      4.1.4 The effect of crosslinking agent on swelling behaviour
      4.1.5 Fourier Transform Infra-Red Analysis
      4.1.6 Thermogravimetric Analysis
      4.1.7 Differential Scanning Calorimetry Analysis
      4.1.8 Morphology study
   4.2 Preparation of hydrogels by electron beam radiation
      4.2.1 Effect of irradiation dose on hydrogels
      4.2.2 Effect of different composition
   4.3 Preparation of optimization hydrogels by ultra violet radiation
      4.3.1 Effect of irradiation time on gel fraction
      4.3.2 Effect of % photo initiator on gel fraction
      4.3.3 Effect of irradiation time on swelling behaviour
      4.3.4 Effect of % photoinitiator on swelling behaviour
      4.3.5 Fourier Transform Infra Red Analysis
      4.3.6 Thermogravimetric Analysis
      4.3.7 Differential Scanning Calorimetry Analysis
      4.3.8 Morphology study
   4.4 Comparison based on morphology study of hydrogels by Electron Beam radiation and Ultra-Violet radiation

5. CONCLUSIONS AND RECOMMENDATION FOR FURTHER STUDY
   5.1 Conclusions
   5.2 Recommendation for further study

REFERENCES
BIODATA OF STUDENT