Cairo University

OPTIMIZATION OF MICROWAVE ASSISTED EXTRACTION OF BETA CAROTENE FROM CARROT PEELS USING RESPONSE SURFACE METHODOLOGY

By

Amira Khaled Anwar Abo El-Ata

A Thesis Submitted to the
Faculty of Engineering at Cairo University
in Partial Fulfillment of the
Requirements for the Degree of
DOCTOR OF PHILOSOPHY
in
CHEMICAL ENGINEERING

FACULTY OF ENGINEERING, CAIRO UNIVERSITY
GIZA, EGYPT
2018

OPTIMIZATION OFMICROWAVE ASSISTED EXTRACTION OF BETA CAROTENE FROM CARROT PEELS USING RESPONSE SURFACE METHODOLOGY

By

Amira Khaled Anwar Abo El-Ata

A Thesis Submitted to the
Faculty of Engineering at Cairo University
in Partial Fulfillment of the
Requirements for the Degree of
DOCTOR OF PHILOSOPHY
in
CHEMICAL ENGINEERING

Under the Supervision of

Prof. Dr.
Mahmoud Abdel Hakim El-Rifai

Professor of Chemical Engineering
Chemical Engineering Department
Faculty of Engineering
Cairo University

Prof. Dr.
Reem Sayed Ettouney

Professor of Chemical Engineering
Chemical Engineering Department
Faculty of Engineering, Cairo University

Dr.
Ayat Osama Ghalab

Associate Professor of Chemical Engineering
Chemical Engineering Department
Faculty of Engineering, Cairo University

FACULTY OF ENGINEERING, CAIRO UNIVERSITY
GIZA, EGYPT
2018

OPTIMIZATION OF MICROWAVE ASSISTED EXTRACTION OF BETA CAROTENE FROM CARROT PEELS USING RESPONSE SURFACE METHODOLOGY

By

Amira Khaled Anwar Abo El-Ata

A Thesis Submitted to the
Faculty of Engineering at Cairo University
in Partial Fulfillment of the
Requirements for the Degree of
DOCTOR OF PHILOSOPHY
in
CHEMICAL ENGINEERING

Approved by the
Examining Committee

____
Prof. Dr. Mahmoud Abdel Hakim El-Rifai, Thesis Main Advisor
Professor, Chem. Eng. Dept., Fac. Of Eng., Cairo University

____
Prof. Dr. Reem Sayed Ettouney, Advisor
Professor, Chem. Eng. Dept., Fac. Of Eng., Cairo University

____
Prof. Dr. Magdi Fouad Abadir, Internal Examiner
Professor, Chem. Eng. Dept., Fac. Of Eng., Cairo University

____
Prof. Dr. Abdel Ghani Gamal Abou El Nour, External Examiner
Professor of Chemical Engineering, Pilot Plant and Chemical Engineering Department, National Research Center, Dokki

FACULTY OF ENGINEERING, CAIRO UNIVERSITY
GIZA, EGYPT
2018

Engineer's Name: Amira Khaled Anwar Abo El-Ata
Date of Birth: 20/5/1986
Nationality: Egyptian
E-mail: Amira_aboelata@yahoo.com
Phone: 01010080545
Address: Maadi, Cairo, Egypt
Registration Date: 1/10/2011
Awarding Date: / / 2018
Degree: Doctor of Philosophy
Department: Chemical Engineering

Supervisors:

Prof. Mahmoud Abel Hakim El-Rifai
Prof. Reem Sayed Ettouney
Dr. Ayat Osama Ghallab

Examiners:

Prof. Mahmoud Abdel Hakim El-Rifai (Thesis main advisor)
Prof. Reem Sayed Ettouney (Advisor)
Prof. Magdi Fouad Abadir (Internal examiner)
Prof. Abel Ghani Gamal Abou El Nour (External examiner)

Title of Thesis:

Optimization of Microwave Assisted Extraction of Beta Carotene From Carrot Peels Using Response Surface Methodology

Key Words:
Response Surface methodology, Microwave assisted Extraction, β-carotene; Carrot peels

Summary:

This thesis presents a detailed response surface methodology study on the microwave assisted extraction of β-carotene from carrot peels. Unlike published research which utilized a modified domestic microwave oven, in this study, a controlled temperature microwave oven was used. Preliminary trials were applied to find the best heating procedure, then a full response surface methodology starting from the first order model passing by the steepest ascent line to the second order model was applied. Optimum conditions were determined and compared with published experimental results.

Acknowledgments

First I thank **ALLAH**, the most gracious, the ever merciful for helping me finishing this work.

I want to thank all those, who helped me by their knowledge and experience by expressing my deepest gratitude to my main supervisor; Professor Mahmoud A. El-Rifai for his continuous support, encouragement, immense knowledge and patient guidance, his guidance helped me in all time of research and writing this thesis.

I am also indebted to my co-supervisor Professor Reem S. Ettouney for her continuous and unconditional help, I owe her for valuable supervision, continuous encouragement, and useful suggestions during this work. My profound thanks are also due to Dr. Ayat O. Ghallab, co-supervisor of my thesis for her continuous support.

No words are enough to express my gratitude to Professor Kamal Youssef, Faculty of Science, Chemistry Department, Minya University for his advice and support during conducting the experimental work in his laboratory.

Last but not the least; my sincere appreciation and profound gratitude to my beloved family for their continuous help and support in not only this work but through my whole life.

Table of Contents

Contents

List of Tables

List of Figures

List of Symbols, Abbreviations, and Nomenclature

ANOVA	Analysis of Variance
C	Hexane composition (v/v)
D	True diffusivity
$\mathbf{D_{eff}}$	Effective diffusivity
ε	Permittivity
$\boldsymbol{\varepsilon_0}$	Permittivity of free space
$\boldsymbol{\varepsilon'}$	Dielectric constant
$\boldsymbol{\varepsilon''}$	Dielectric loss
FMASE	Focused microwave assisted soxhlet extraction
$\mathbf{K_D}$	Distribution coefficient
L/S	Liquid to solid ratio, ml/g
MAE	Microwave Assisted Extraction
PLE	Pressurized Liquid Extraction
RSM	Response Surface Methodology
$\boldsymbol{tan\ \delta}$	Dissipation factor
UAE	Ultrasound Assisted Extraction
$\mathbf{X_1}$	Coded value for L/S ratio
$\mathbf{X_2}$	Coded value for hexane composition
Y	β-Carotene yield, mg/100 g peels
$\boldsymbol{\phi}$	Fraction of space available for extracting solvent
γ	Tortuosity factor

Abstract

This thesis presents a detailed study on the microwave assisted extraction of β-carotene from carrot peels as a modern technique compared to conventional methods such as soxhlet extraction. The published literature covering microwave assisted extraction in different fields was based on the use of a modified domestic microwave oven which has its own drawbacks as it lacks control of temperature. In this study, a controlled temperature microwave oven was used. This relaxes the necessity of following up the effect of the set microwave power on the temperature evolution.

Preliminary trials were conducted to select the best heating procedure along with the appropriate values for the rest of the parameters to start investigating around. A factorial design was chosen to conduct the trials where a response surface methodology (RSM) is used to analyze obtained experimental data with a view to maximizing the β-carotene yield obtainable from local carrot peels. The design of the experiments has been conceived to delineate the effects of preheating the solvent before MAE, solvent composition, liquid to solid (L/S) ratio, and extraction time on the obtainable yield.

Unlike conventional techniques where any solvent can heat up, only polar/ionic solvents can respond to microwave irradiation, that is why a mixture of hexane and ethanol is used as the solvent in order to combine good dissolution and high absorbance to microwave energy. Though acetone alone was conventionally used in earlier similar studies, it was excluded in this study since it is a toxic solvent and not suitable for food grade β-carotene. The maximum extraction temperature set on the microwave oven was 50°C. Solvent preheat proved beneficial in reducing the MAE time since it provides for the latent heat of vaporization of the water-hexane azeotrope evolved on extraction.

The application of RSM starting from the first order model passing by the steepest ascent line to the second order model enabled to determine the optimum operating conditions of 5 minutes preheating time, 2 minutes extraction time, a solvent composition of 0.747 (v/v) hexane, and a (L/S) ratio of 61.08 ml/g. The maximum yield of 141.77 mg β-carotene per 100 g peels was obtained. Comparing obtained results with published literature utilizing the same technique was very promising; this study resulted in a higher yield with a lower L/S ratio and less extraction time which is attributed to the suggested solvent preheating technique.

Chapter 1: Introduction

Solid liquid extraction is resorted to in many applications covering chemical processing, pharmaceutical industries, and qualitative and quantitative analyses. A wide variety of conventional contacting equipment has been used such as soxhlet, automated soxhlet, and high pressure soxhlet extraction. New techniques are under experimental studies and proved superiority in reducing both extraction time and the amount of solvent. Focused microwave assisted soxhlet extraction, microwave assisted extraction, and ultrasound assisted extraction are examples of such techniques. This thesis presents a study on the microwave assisted extraction of β-carotene from local carrot peels using a preheating technique.

Chapter (2) starts by giving a brief introduction on carotenoids and their subtypes: Carotenes and Xanthophylls. Carotenoids physical and chemical properties are reviewed, and also natural and synthetic sources are mentioned. The growing uses of carotene and its reflection on the market share are discussed. Different techniques of carotene extraction are presented including the use of super critical fluids, micro emulsions and pressurized fluids. This chapter also presents a detailed comparison between the different equipment used in the solid liquid extraction. The solid-liquid diffusion theory is then explained in general cases and in the specific cases of the Dunaliella salina algae which is the highest precursor of β-carotene among all sources. Finally details related to microwave heating are discussed starting from the theory of microwave energy passing by its various applications in different fields focusing on the microwave assisted extraction of carotenes from carrots, carrot peels and algae.

Chapter (3) outlines the problem to be addressed in this work where a lab scale controlled temperature microwave oven is used to maximize β-carotene yield obtained from local carrot peels using a safe food grade solvent. The analysis of the experimental data will be carried out using response surface methodology with a view to obtain the optimum extraction conditions.

Chapter (4) presents the experimental methodology. The first part outlines the steps taken in the design of the experiment, the design used, and the path from the initial conditions up to the optimized ones. The second part details the different parameters to be considered, their proper preliminary value selection and the selection of the parameters to be optimized. These include different categories such as parameters related to the amount of heat supplied to the material (microwave power, temperature, and extraction time), physical and chemical properties of the material under extraction (pretreatment methods and contact surface area), and used solvent (chemical structure and its ratio to the sample used).

Chapter (5) details the experimental methodology. It starts by discussing the criteria of selection of the natural raw material subjected to the extraction process, and the properties of the chemicals used. Detailed explanation of all equipment used in this study is also presented. Finally the procedure undertaken in each trial starting from the material preparation passing by the extraction process and ending by the analysis is explained.

Chapter (6) presents the results of the conducted experimental work. Results of preliminary trials conducted to select the best heating procedure along with the appropriate values for the rest of the parameters to start investigating around are first presented. Solvent preheat proved beneficial in reducing the MAE time. A detailed response surface methodology was conducted where a first order model describing the first phase of the experiment was conducted followed by the second phase which included the path along the steepest ascent line until reaching the third phase where a second order model was conducted. Finally the optimum conditions were calculated and compared to published literature. The results of this study showed superiority with respect to yield with a lower L/S ratio and less extraction time. This is attributed to the suggested solvent preheating technique.

Chapter (7) summarizes the main conclusions reached in this work.