HEAT AND MICROWAVE REFLUX EXTRACTION, OPTIMIZATION, AND PHYSICOCHEMICAL CHARACTERIZATION OF OLEORESINS FROM MALAYSIAN PEPPER (*Piper nigrum*)

OLALERE, OLUSEGUN ABAYOMI

Doctor of Philosophy

UNIVERSITI MALAYSIA PAHANG

SUPERVISOR'S DECLARATION

We hereby declare that we have checked this thesis and in our opinion, this thesis is adequate in terms of scope and quality for the award of the degree of Doctor of Philosophy

_____

(Supervisor's Signature)

Full Name: PROF. DR ABDURAHMAN HAMID NOUR

Position: PROFESSOR

Date: 1st August 2018

_____

(Co-supervisor's Signature)

Full Name: PROF. DATO' DR ROSLI BIN MOHD YUNUS

Position: PROFESSOR

Date: 1st August 2018

STUDENT'S DECLARATION

I hereby declare that the work in this thesis is based on my original work except for quotations and citations which have been duly acknowledged. I also declare that it has not been previously or concurrently submitted for any other degree at Universiti Malaysia Pahang or any other institutions.

_____

(Student's Signature)

Full Name: OLALERE, OLUSEGUN ABAYOMI

ID Number: PKC15013

Date: 1st August 2018

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LIST OF SYMBOLS

h_1	Extraction time in heat reflux extraction
h_2	Feed particle size in heat reflux extraction
h_3	Feed-solvent ratio in heat reflux extraction
x_1	Irradiation time in microwave refluxation
x_2	Microwave power in microwave refluxation
x_3	Feed particle size in microwave refluxation
x_4	Feed-solvent ratio in microwave refluxation
y'_w	Extraction yield from heat refluxed white pepper
y'_b	Extraction yield from heat refluxed black pepper
y_w	Extraction yield from white pepper microwave refluxation
y_b	Extraction yield from black pepper microwave refluxation
$y'_{w(av)}$	Average extraction yield from heat refluxed white pepper
$y'_{b(av)}$	Average extraction yield from heat refluxed black pepper
$y_{w(av)}$	Average extraction yield from white pepper refluxation
$y_{b(av)}$	Average extraction yield from black pepper refluxation
$Q_t'_w$	Absorbed energy by heat refluxed white pepper sample
$Q_t'_b$	Absorbed energy by heat refluxed black pepper sample
$Q_{t\ w}$	Absorbed microwave energy by white pepper sample
$Q_{t\ b}$	Absorbed microwave energy by black pepper sample
$Q_t'_{w(av)}$	Average absorbed energy by heat refluxed white pepper sample
$Q_t'_{b(av)}$	Average absorbed energy by heat refluxed black pepper sample
$Q_{t\ w(av)}$	Average absorbed microwave energy by white pepper sample
$Q_{t\ b(av)}$	Average absorbed microwave energy by black pepper sample
a	Values are means ±SD of triplicate runs

LIST OF ABBREVIATIONS

ANOM	Analysis of Mean
ARP	Antiradical Power
BPOE	Black Pepper Oleoresin Extracts
HRE	Heat Reflux Extraction
MRE	Microwave Reflux Extraction
PI	Performance Index
REI	Relative Extraction Index
SFE	Single Factor Experiment
SNR	Signal to noise ratio
TODOE	Taguchi orthogonal design of experiment
WPOE	White Pepper Oleoresin Extracts

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ABSTRAK

Terdapat pelbagai bukti yang mendedahkan keupayaan tumbuhan berfungsi yang tidak dapat ditandingi dalam mengubati penyakit berpunca dari radikal bebas degeneratif dalam bidang etno-perubatan. Lada hitam dan putih (Piper nigrum) adalah salah satu contoh tumbuhan berfungsi yang kaya dengan ciri-ciri perubatan dan nutrisi berikutan keupaayan untuk menghalang atau memusnahkan pembentukan radikal bebas. Ciri penting ini diperoleh tanaman komoditi utama tersebut berikutan kandungan pelbagai jenis unsur mineral serta sebatian fenolik di dalamnya.Justeru itu, satu keperluan kemudiannya timbul untuk mengenal pasti teknik pengekstrakan yang cekap, di mana ekstrak fenolik yang berkualiti tinggi pada keadaan optimum dapat diperolehi. Berikutan itu, dalam kajian ini,teknik-teknik pengekstrakan haba refluks (HRE) dan pengekstrakan mikro refluks (MRE) dibandingkan untuk pengekstrakan oleoresins daripada lada hitam dan putih. Kesan kaedah pengekstrakan pada hasil pengekstrakan, komposisi unsur, kandungan fenolik, aktiviti antioksidan, mikrostruktur dan ciri-ciri berfungsi telah dikaji dengan ringkas dan jelas. Seterusnya, prestasi ataupun keberkesanan kedua-dua teknik pengekstrakan ini dinilai pada tahap operasi yang berbeza. Sehubungan dengan itu, kesan parameter yang berbeza terhadap hasil pengekstrakan dan jumlah tenaga terserap telah dikaji dengan mengaplikasikan eksperimen faktor tunggal (SFE) dan Taguchi reka bentuk ortogon eksperimen (TODOE). Keputusan yang diperolehi menunjukkan bahawa MRE memberikan ekstrak yang lebih baik daripada HRE dari segi minyak tetap yang diekstrak, jumlah kandungan fenolik, kapasiti antioksidan serta bilangan sebatian fenolik dan unsur-unsur mineral. Indeks prestasi anggaran pula mengesahkan bahawa kaedah MRE adalah empat kali dan enam kali lebih baik daripada HRE, masing-masing dalam pengrefluksan lada hitam dan putih. Tambahan pula, lada putih menawarkan antioksidan semulajadi yang lebih baik dan berkualiti dari segi kuantiti minyak tetap, jumlah kandungan fenolik, kapasiti antioksidan serta bilangan sebatian fenolik dan unsur-unsur mineral. Oleh itu, kajian ini member suatu konsepsi optimum dari aspek kualitatif dan kuantitatif mengenai kesan teknik pengekstrakan konvensional dan bukan konvensional pada hasil, komposisi mineral, kandungan fenolik, aktiviti antioksidan, ciri-ciri morfologi dan berfungsi bagi ekstrak lada hitam dan putih secara terperinci.

ABSTRACT

Emerging evidence has revealed the undeniable ability of functional plants for the treatment of many degenerative free radical-related diseases in ethnomedicine. Black and white peppers (*Piper nigrum*) are examples of such plants with both nutritional and medicinal properties. The free radical scavenging properties of these important commodity crops is due to the presence of various mineral elements and phenolic compounds. A need then arises to find an efficient extraction technique that gives high quality phenolic extracts at optimum condition. In this study, heat reflux extraction (HRE) and microwave reflux extraction (MRE) techniques were compared for the extraction of oleoresins from black and white pepper. The effects of these extraction methods on the extraction yield, elemental composition, phenolic content, antioxidant activities, microstructure, and functional characteristics were succinctly investigated. The performance of the two extraction techniques were thereafter evaluated at different operating levels. This was achieved firstly by examining the effects of different parameters on both the extraction yield and absorbed energy, using a single factor experiment (SFE) and Taguchi orthogonal design of experiment (TODOE). The result obtained revealed that the MRE presented a better extracts than HRE in terms of fixed oil extracted, total phenolic content, antioxidant capacity, number of phenolic compounds and mineral elements. The estimated performance index adjudged MRE to be four times and six times better than HRE in black and white pepper refluxation, respectively. Moreover, white pepper offered a better and quality natural antioxidants in terms of the quantity of fixed oil, total phenolic content, antioxidant capacity, number of phenolic compounds and mineral elements. This research therefore provided an optimized, qualitative and quantitative insight into the effects of conventional and non-conventional extraction techniques on the yield, mineral composition, phenolic content, antioxidant activities, morphological and functional characteristics of black and white pepper extracts.

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