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HERBS RECOGNITION SYSTEM BASED ON PHYSIOCHEMICAL PROPERTIES USING WEIGHTED HISTOGRAM AND MULTIPLE DISCRIMINANT ANALYSIS METHODS

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By

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Currently, herbs recognition system has become a promising method to identify herbs species. Misuse of herbal medicine can cause serious health problems due to toxicological effects of phytochemical. As a result, a system that able to distinguish the types of herbs is needed. Most herbs recognition systems available in the market are dependent on experts. In this research, the concern is to identify the herbs compounds within the same group species where the physical appearance and aroma are similar. The work mainly focuses on herbs recognition systems that intended for researchers and medical practitioners use without the need for experts.

The study is mainly based on chemical and physical properties as well as the combination of both. Hence, three feature extraction methods for herbs recognition system based on chemical and physical properties of herbs are presented. Electronic Nose (E-Nose) devices have been used extensively to differentiate and characterize the herb species based on their unique odor. Electrical signal generated from the gas sensor array is one of the physical properties studied. Then, a Gas Chromatography-Mass Spectrometry (GCMS) device is utilized to extract the chemical compound of herbs. The first feature extraction technique in this research is Principal Component Analysis (PCA) and selected feature based on electrical signal, however it is an unsupervised learning. Thus, Multiple Discriminant Analysis (MDA) is proposed as the second feature extraction technique. MDA is one of the supervised learning techniques to replace PCA. The third feature extraction technique is proposed to develop an automated GCMS system that differentiates the herbs species from the major volatile compounds. The Weighted Histogram Analysis Method (WHAM) is

proposed to make use of both major and minor volatile compounds in GCMS herbs recognition system.

Fusion techniques have been extensively studied on multisensory environments. Comparison between system with and without feature fusion techniques is presented. In this research, 19 herbs species from 5 family groups are studied. The robustness test of the three proposed herbs recognition systems are performed via four classification models: Support Vector Machine (SVM), K-Nearest Neighbors (KNN), Multinomial Logistic Regression (MLR), and Gaussian Radial Basis Function (RBF) Kernel. In GCMS, the overall system performance with WHAM improves the classification accuracy by average of 0.6%-38.34% using SVM and KNN. In E-Nose herbs recognition system, an average between 0%-21.73% improvement in system performance with MDA. The performance of classification accuracy using KNN shows a better result within the family group from 92.15% to 100% compared to the other methods. In addition, KNN method shows classification accuracy improvement in average of 99.58%-100% within the same family group.

As a conclusion, the system with KNN method is capable to classify herbs species more accurately as compared to the other three classification method. The innovation of this research could benefit especially the researchers, to identify the plant species without relying on the expertise of botanists and forest rangers for the learning and training process.

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SISTEM PENGECAMAN HERBA BERDASARKAN CIRI FIZIKIMIA MENGGUNAKAN KAEDAH ANALISIS HISTOGRAM BERAT DAN ANALISIS PELBAGAI DISKRIMINASI

Oleh

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Pada masa kini, sistem pengecaman herba telah menjadi kaedah yang berkemampuan untuk mengenal pasti spesies herba. Penyalahgunaan ubat herba boleh menyebabkan masalah kesihatan yang serius akibat kesan daripada toksikologi fitokimia. Oleh itu, sistem yang dapat membezakan jenis herba amat diperlukan. Kebanyakan sistem pengecaman herba yang terdapat di pasaran memerlukan kebergantungan pakar. Dalam penyelidikan ini, keutamaan dalam mengenal pasti sebatian herba dalam kumpulan spesies yang sama di mana rupa fizikal dan aroma adalah hampir serupa. Kerja ini tertumpu kepada sistem pengecaman herba yang bertujuan untuk kegunakan penyelidik dan pengamal perubatan tanpa memerlukan khidmat pakar.

Kajian utama adalah berdasarkan sifat kimia dan sifat fizikal dan beserta gabungan dari kedua-duanya. Oleh itu, tiga kaedah pengekstrakan ciri untuk sistem pengecaman herba berdasarkan sifat kimia dan sifat fizikal herba dibentangkan. Peranti Hidung Elektronik (E-Nose) telah digunakan secara meluas bagi membezakan spesies herba berdasarkan bau uniknya. Isyarat elektrik yang dihasilkan daripada sensor-sensor gas adalah salah satu sifat fizikal yang dikaji. Kemudian, Gas Kromatografi-Spektrometri Mass (GCMS) digunakan untuk mengekstrak sebatian kimia herba. Teknik pengekstrakan ciri pertama dalam penyelidikan ini ialah Analisis Komponen Utama (PCA) dan ciri terpilih adalah berdasarkan isyarat elektrik, namun teknik ini merupakan pembelajaran unsupervised. Oleh itu, Analisis pelbagai diskriminasi (MDA) dicadangkan sebagai teknik pengekstrakan ciri kedua. MDA adalah salah satu teknik pembelajaran supervised yang digunakan untuk menggantikan PCA. Teknik pengekstrakan ciri yang ketiga dicadangkan adalah menghasilkan sistem

GCMS secara automatik yang dapat membezakan spesies herba daripada sebatian meruap. Kaedah Analisis Histogram Berat (WHAM) dicadangkan untuk membolehkan kedua-dua sebatian meruap major dan minor dalam sistem pengecaman herba GCMS.

Teknik gabungan telah dikaji secara meluas pada bidang multisensor. Perbandingan antara sistem bersama atau tanpa teknik gabungan ciri telah dibentangkan. Dalam penyelidikan ini, 19 spesies herba daripada 5 kumpulan keluarga dikaji. Ujian keteguhan keatas ketiga-tiga sistem pengecaman herba yang dicadangkan telah dilakukan melalui empat jenis model klasifikasi: Mesin Vektor Sokongan (SVM), K-Jiran Terdekat (KNN), Regresi Logistik Multinomial (MLR), and Kernel Fungsi Asas Jejari Gaussian (RBF). Dalam GCMS, keseluruhan prestasi sistem beserta WHAM telah meningkatkan ketepatan klasifikasi dengan purata 0.6%-38.34% dengan menggunakan SVM dan KNN. Dalam sistem pengecaman herba E-Nose, purata antara 0%-21.73% peningkatan dalam prestasi sistem beserta MDA. Prestasi ketepatan pengelasan menggunakan KNN menunjukkan hasil yang lebih baik dalam kumpulan keluarga daripada 92.15% hingga 100% berbanding kaedah-kaedah yang lain. Di samping itu, kaedah KNN menunjukkan peningkatan ketepatan klasifikasi secara purata 99.58%-100% dalam kumpulan keluarga yang sama.

Kesimpulannya, sistem dengan kaedah KNN mampu mengklasifikasikan spesies herba dengan lebih tepat berbanding tiga kaedah pengelasan yang lain. Inovasi penyelidikan ini boleh memberi manfaat terutamanya kepada penyelidik, untuk mengenal pasti spesies tumbuhan tanpa bergantung kepada kepakaran ahli botani dan renjer hutan untuk proses pembelajaran dan latihan.

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Declaration by Members of Supervisory Committee

This is to confirm that:

- the research conducted and the writing of this thesis was under our supervision;
- supervision responsibilities as stated in the Universiti Putra Malaysia (Graduate Studies) Rules 2003 (Revision 2012-2013) are adhered to.

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LIST OF ABBREVIATIONS

AEC	Amperometric Electrochemical
ANFIS	Adaptive Neuro-Fuzzy Inference System
ANN	Artificial Neural Network
ANOVA	Analysis of Variance
CAPSI	Computer Aided Plant Species Identification
CP	Conducting Polymers
CV	Cross Validation
E-Nose	Electronic Nose
E-Tongue	Electronic Tongue
EI	Electron Ionisation
EMC	Eigenspace Method based on Class features
Gaussian RBF Kernel	Gaussian Radial Basis Function Kernel
GC	Gas Chromatography
GCMS	Gas Chromatography Mass Spectrometry
GLCM	Gray-Level Co-occurrence Matrix
IBS	Institute of Bioscience
KNN	K-Nearest Neighbors
LDA	Linear Discriminant Analysis
MDA	Multiple Discriminant Analysis
MLP	Multi Layer Perceptron
MLR	Multinomial Logistic Regression
MOS	Metal Oxide Semiconductor
MS	Mass Spectrometry
PCA	Principal Component Analysis

QCM	Quartz crystal microbalance
RTIC	Reconstructed Total Ion Current
SAW	Surface acoustic wave
SVM	Support Vector Machine
UPM	Universiti Putra Malaysia
UV	Ultra Violet
VOCs	Volatile Organic Compounds
WHAM	Weighted Histogram Analysis Method

CHAPTER 1

INTRODUCTION

1.1 Research background

Herb is a unique plant species. Every part in herbs is very useful as it provides significant impact in various fields including medicine, food industry, healthcare product, culinary, and pharmaceutical. Herbs are rich in pythochemicals. Phenolic compounds (phenolic acids, flavonoids, terpenes etc.) are the main phytochemical which have powerful antioxidant activity in herbs (Saranraj, et al., 2019). However, its usage without appropriate study may cause negative side effects on one's health due to lack of information, resources and expertise in identifying herb species. Overconsumption of herbs may influence human health due to toxicological effects of phytochemical. Many scientific literatures focus on psychoactive herbal extracts and their pythochemicals to identify herbs species (Ayad and Akkal, 2019; Graziano *et al.*, 2017; Ramirez-Marroquin and Jimenez-Arellanes, 2019; Umit Sayin, 2016).

During the past decades, the interest in phytochemical studies has increased following serious health problems due to misuse of herbal medicine and it is a worldwide issue. A lot of plant species have been used for medical purposes (Lawal *et al.*, 2019; Nagalingam, 2017; Sikarwar, 2017; Yuan *et al.*, 2016). Although the traditional plant-based (herbal) medicines are natural products, the use of herbs in medicinal products and supplements without systematic observation can cause serious adverse reactions (Ekor, 2014). Numerous cases of herbal toxicity and herbal drug reaction have been reported (Kahraman *et al.*, 2020; Nudrat and Naira, 2016; Vidushi, 2013). This is due to lack of information, unqualified practitioners and inadequate data about the herbs.

In August 1995, the first version of herbal website - Henriette's Herbal Homepage, has been published online (Dighe, 2010). The databases are to provide reference for botanical works. However, there are still nearly one-fifth of plants left to discover, not including the undisclosed plants. Around 2,000 new plants worldwide were discovered every year. Yet, numerous plants species are still unknown (Antonelli *et al.*, 2020; Cheek *et al.*, 2020).

Deep researches on chemical compounds that may come from leaves, stems, flowers, roots, and seeds are needed (Djamil *et al.*, 2021; Gaca *et al.*, 2021; Sun *et al.*, 2021; Zhang *et al.*, 2021). Due to the complex mixtures of chemical compounds, researchers such as botanist, forest ranger and scientist require a powerful analytical chemistry tool to identify and quantify the compounds present in the herbs. Nowadays, several chromatography technologies have been invented to separate the chemical mixture from solid or fluid sample such as Gas

Chromatography Mass Spectrometry (GCMS) (Mehdi *et al.*, 2021), Liquid Chromatography Mass Spectrometry (LCMS) (Iwan *et al*., 2021) and High Performance Liquid Chromatography (HPLC) (Zaheer *et al.*, 2021). The chemical compounds libraries are pre-installed in these machines. The competence of such machine depends on the variation of mass-spectral libraries available. Even though many countries have utilized these technologies, some institutions cannot afford to have the facility due to the expensive cost of the machine, equipment, services and mass-spectral library. Besides, only experts can conduct the experiments.

To overcome these problems, scientists and engineers have designed many intelligent solutions such as electronic sensing instruments - Electronic Nose (E-Nose) (Elham *et al.*, 2021; Lucas *et al.*, 2021; Wijaya *et al.*, 2021; Xu *et al.*, 2021) and Electronic Tongue (E-Tongue) (Ross *et al.*, 2021; Wang *et al.*, 2021; Zaukuu *et al.*, 2021), by using gas sensors and taste sensors array respectively. Using a cost-effective way to develop the system, it helps many institutions to self-conduct the experiment for various purposes without the need for experts. These devices are small in size, enabling the users to carry them and operate them in real time on site easily. This is especially an advantage for forest rangers. Since these applications consist of a group of sensors, the disadvantage of this technology is the limited number of available sensor types suitable for herbs. Besides, gas sensor is highly sensitive to humidity and temperature. While some sensors may have short lifetime. For example, the life expectancy of a gas sensor is about 6 months.

Both chemical and physical system approaches promise good classification for certain herbs. However, the critical problem in this research is to identify the herbs compounds within the same group species where the physical appearance and aroma are similar. By taking the pros and cons of these two technologies, a feature-fusion technique from both sources can help achieve improved classification as compared to single modality. The investigation on the classification performance among the three different methods will be the main focus of this research.

1.2 Problem Statement

Each plant species has a unique physicochemical and distinctive odor. Odor is one of the most important indicators to distinguish the herbs species. The challenge in identifying the species is, it is hard to differentiate herbs under same family group species since the characteristics and aromas are almost identical. Two instrumentation technologies have been invented to analyze the physical and chemical properties of herbs odor using E-Nose and GCMS.

The invention of E-Nose technologies for detection and identification has accelerated rapidly since the 1980s (Persaud and Dodd, 1982). Selection of suitable gas sensors to detect the gas released from herbs and the design of pattern recognition system are two important components in E-Nose development used to identify the complex odor of herbs. The implementation of many gas sensors array in E-Nose device can benefit the system by deriving more electrical signal to be extracted in order to understand the herbs better. However, it tends to have a high-dimensional data. A dataset with high dimensionality may lead to the curse of dimensionality problem. Most research uses Principal Component Analysis (PCA) to mitigate the problems associated with high dimensional data (Jolliffe and Cadima, 2016; Mishra, 2017). This method is promising in reducing the data dimension by orthogonal transformation but at some point, it is an unsupervised learning. It only focuses on projecting all data points to the axes with maximum variance without separating the class species. Unlike Multiple Discriminant Analysis (MDA), it is another dimensional reduction method. The difference between MDA and PCA is, MDA is a supervised data dimension reduction method. Data will be projected to the dimension that perform high separation between classes and low separation within classes.

The capability of headspace GCMS technique to analyze the volatile compounds by extracting the chromatographic signal and identify the chemical components has been used for decades. In practical situation, the pattern of major chemical compounds helps herbs expert such as botanists, forest rangers, and scientist to identify the species. However, this is a manual herbs recognition process. No automated herbs recognition system has been applied into GCMS technology. Without neglecting the minor signal in the GCMS, herbs recognition system tends to cause a bias distribution of chromatographic signals. Therefore, Weighted Histogram Analysis Method (WHAM) technique has been proposed for extracting new features from minor and major volatile compound data of chemical properties. WHAM discovered that using multiple histogram weighting technique allowed it to extract all data at once while also reducing the dimensionality of data features.

Fusion techniques have been greatly used on multisensor environments. The aim is to improve system performance. The framework of feature fusion herbs recognition system was inspired by many past researches on the fusion on E-Nose and Electronic Tongue (Haddi *et al.*, 2013; Haddi *et al.*, 2014; Men *et al.*, 2014; Wang *et al.*, 2021). However, in most studies, fusion data are from the same modality (physical properties). Therefore, a new approach of using data from two modalities (chemical and physical properties) was proposed. The proposed algorithm gives a huge potential in herbs recognition system to overcome the problem of single modality system that only depends on one source, which is chemical or physical property.

1.3 Objectives

The main objective of this research is to design and develop three herbs recognition system based on physical and chemical properties (E-Nose herbs recognition system, GCMS recognition system, and feature fusion herbs recognition system). In addition, the research also deals with the following sub-objectives:

1. To propose an automated system in GCMS herbs recognition system and introducing WHAM for feature extraction.
2. To improve the accuracy of E-Nose herbs recognition system by incorporating MDA method.
3. To improve overall system by proposing feature fusion technique by combining two modalities which are chemical and physical properties.
4. To evaluate and testing of the proposed method.

1.4 Scope of Research

The scope of this research includes the following:

1. Five family of aromatic herbs species were chosen from a hundred along with the relevant subspecies (19 species in total).
2. All leaves samples were plucked from trees at the same location, the conservation park at Institute of Bioscience (IBS), Universiti Putra Malaysia, to ensure standardized constant factors such as nutrition soil, climate, biosis and growth regulators.
3. Mature leaves were used in this research to get stable volatile compound. To capture maximum volatile compound, it is important to pluck the herb leaf early in the morning to keep highest level of freshness. Botanist from IBS had helped in validating the mature leaves.
4. Headspace GCMS technique was applied in this study. The advantage of headspace technique as compared to essential oil is that, the analysis can be carried out in less time and it requires a small number of leaves for one sample.
5. The metal oxide semiconductor gas sensors were used for E-nose system.
6. All the findings for three proposed herbs recognition systems are based on the five group species only.

1.5 Research Contributions

The contributions in this research are listed as follows:

1. Developing a herbs recognition system based on chemical properties and physical properties to identify the herbs species in the same family. This work provides 3 different herbs recognition systems to identify the herbs species. The herbs recognition system with single modality only uses data from the chemical properties or physical properties. In this research, GCMS and E-Nose are considered as the single modality systems. In this research, new databases of herbs species based on chemical properties (GCMS) and physical properties (E-Nose) have been developed using these two herbs recognition systems. This new

system that combines both properties is considered as the multiple modalities system. The performance of these 3 different herbs recognition systems were evaluated based on the accuracy of each system to classify the herbs species.

2. Providing a new feature extraction method based on chemical properties using Weighted Histogram Analysis Method (WHAM).
Implementation of pattern recognition system into GCMS system will benefit the herbs recognition process with no botanist required. Building this automated system from GCMS signal is complicated because the data is represented in discrete numbers of volatile organic compounds (VOCs). The formulation of new feature extraction using WHAM makes it possible to transform the data into a graphical representation by weighting multiple single features histogram. The proposed method offered the ability to gather more information from major and minor VOCs by employing correlation between features from two single histograms of VOCs peak area and height. Besides, this method of WHAM can produce a unique pattern of correlation histogram which can be used in developing a new herbal database.

3. Providing a new feature fusion technique by combining two different modalities which are chemical properties and physical properties.
The research work focuses on recognizing the herbs species from the same family group. The critical parameter that needs to be explored, is when the herbs under the same family has high possibility of having almost similar physical appearances and aroma characteristics. The similarity of aroma indicates the similar signal pattern of chromatography in herbs chemical compound. The formulation of the new feature fusion technique gives the best solutions in classifying herbs species based on both properties. The implementation of feature fusion technique for different modalities is a relatively new approach for herbs recognition system.

1.6 Thesis Outline

The thesis is divided into five chapters:

- Chapter 1 introduces the thesis which covers some background information on the plant recognition system based on chemical and physical approaches. The problems in the traditional plant recognition system based on both approaches have been highlighted and identified as the key point for the research. The chapter also consists of the research scope, objectives, research contribution and layout of the thesis.

- Chapter 2 reviews the past and some of the most recent single modality techniques based on both chemical and physical approaches in plant recognition system through literature search. The alternative techniques of fusion between multiple modalities data are also presented in this chapter. These techniques cover the features fusion, decisions fusion and hybrid fusion. A review on the plant recognition algorithms which elaborates several methods for signal processing, feature selection and extraction technique, dimension reduction, classification methods and process validation is also covered in this chapter.

- Chapter 3 presents an overview of the framework for herbs recognition system using chemical and physical approaches. Discussions and experiments were also carried out based on chemical approach using GCMS and physical approach using E-Nose system. The formulation and combination of these two single modalities using fusion techniques for herbs recognition system were designed. The techniques and algorithms are described throughout the chapter, including pre-processing, feature selection and extraction technique, dimension reduction, classification methods, and process validation.

- Chapter 4 discusses the results of the research and evaluation of the herbs recognition system's performance. Recognition results obtained from the presented methods are reported. A comparison between the obtained results and the analysis of the work conducted by means of the single modality techniques and multiple modalities techniques using fusion techniques are also reported.

- Chapter 5 is the conclusion of this thesis and it includes a general overview of the work done. It also summarizes the achievements and further work recommendation to be carried out.

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