

CHEMOMETRICS FUZZY AUTOCATALYTIC SET METHOD
FOR HALAL AUTHENTICATION OF GELATIN

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DEDICATION

Bismillah.

This thesis is dedicated to my family and friends.

And hopefully the outcome and knowledge in this manuscript may be
beneficial for everyone.

inshaALLAH

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ABSTRACT

Fourier transform infrared (FTIR) spectroscopy is one of the established techniques in food adulteration analysis. The issue of halal authenticity of food and pharmaceutical products has become a concern among Muslims due to fraud and unknown sources of ingredients. The most common non-halal ingredient found in some food and pharmaceuticals products is porcine gelatin. In addition, there are other sources of gelatin such as bovine and fish. Fuzzy autocatalytic set (FACS) is a concept that incorporates fuzziness and autocatalytic set. The study related to the characteristics and uses of FACS method is progressing significantly, especially in the area of chemical engineering and also signal and network analysis. However, in this research, FACS is implemented on FTIR spectra of bovine, porcine, and fish gelatins since their spectra exhibit very similar patterns and thus further analysis is required to differentiate them. The spectra of the gelatin are analyzed using a new developed chemometrics technique, namely chemometrics FACS (c-FACS). Several definitions and theorems are introduced to furnish the c-FACS technique. A coded algorithm called Multisystem Dynamic Identification of Gelatin Sources is built for the technique. The algorithm is executed on the gelatins' spectra. The results showed that each gelatin has a unique range signature of wavenumbers and porcine gelatin particularly showed distinct pattern compared to others. The efficiency and the rigorousness of the c-FACS are established when compared to the principal component analysis (PCA) method.

ABSTRAK

Spektroskopi penjelmaan inframerah Fourier (FTIR) merupakan salah satu kaedah mantap yang diterima pakai dalam analisa pengadukan makanan. Isu kesahihan halal dalam produk makanan dan farmaseutikal telah menjadi perhatian masyarakat Islam kerana penipuan dan ketidakpastian akan sumber di dalam ramuannya. Salah satu bahan tidak halal yang paling biasa ditemui di dalam beberapa produk makanan dan farmaseutikal ialah gelatin bersumberkan khinzir. Selain itu, terdapat juga sumber gelatin yang lain seperti lembu dan ikan. Set autopemangkin kabur (FACS) merupakan konsep yang terhasil dari gabungan konsep kabur dan set autopemangkin. Kajian berkaitan ciri-ciri dan penggunaan kaedah FACS sedang giat dijalankan, terutamanya di dalam bidang kejuruteraan kimia dan juga analisis isyarat dan rangkaian. Walaubagaimanapun, dalam penyelidikan ini, FACS diaplikasikan ke atas spektrum FTIR bagi gelatin bersumberkan bahan dari lembu, khinzir, dan ikan kerana spektrum gelatin-gelatinnya mempamerkan corak yang sama dan oleh itu analisis yang selanjutnya diperlukan untuk membezakannya. Spektrum gelatin-gelatin ini dianalisa menggunakan teknik kimometrik yang baharu dibangunkan yang dinamakan sebagai kimometrik FACS (c-FACS). Beberapa takrifan dan teorem diperkenalkan bagi teknik c-FACS. Satu algoritma berkod disebut sebagai Identifikasi Multisistem Dinamik Bagi Sumber Gelatin telah dibina bagi teknik ini. Algoritma tersebut diimplementasikan pada spektra gelatin. Hasil kajian menunjukkan bahawa setiap gelatin mempunyai tanda gelombang nombornya tersendiri, lebih-lebih lagi gelatin khinzir menunjukkan corak yang berlainan berbanding yang lain. Keberkesanan dan kerapian c-FACS terserlah apabila dibandingkan dengan kaedah analisis komponen prinsipal (PCA).

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LIST OF ABBREVIATIONS

ACS	-	Autocatalytic Set
FACS	-	Fuzzy Autocatalytic Set
FTIR	-	Fourier transform infrared
PCA	-	Principal Component Analysis
PF	-	Perron Frobenius
PFE	-	Perron Frobenius eigenvector

LIST OF SYMBOLS

E	-	Set of edges
V	-	Set of vertices
e_i	-	The i^{th} edge
v_i	-	The i^{th} vertex
A	-	Square Matrix
a_{ij}	-	The entries of square matrix A .
G_F	-	Fuzzy graph
$h(e_i)$	-	Fuzzy head of i^{th} edge
$t(e_i)$	-	Fuzzy tail of i^{th} edge
(i, j)	-	Directed link from node i to node j
λ	-	Eigenvalue
$\mu(e_i)$	-	Membership value of i^{th} edge
\sum	-	Summation of
\mathbb{R}	-	Real number
\mathbb{N}	-	Natural number

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CHAPTER 1

INTRODUCTION

1.1 Background of Research

Food and pharmaceutical products are essential needs for human beings. In line with industrialization and globalization, these products have to meet the demand of society along with technological developments in the industries. Some of these products may have been labeled with incorrect information regarding the source of the ingredients (Montowska and Pospiech, 2010). This has become an issue for some religious adherents such as the Muslims, since the status of the raw materials used for the ingredients and the production process may not fulfill their religious requirements.

According to Al-Qaradawi (1994), the Quran has stated the raw materials that are prohibited include pork or flesh of pigs. Gelatin which extracted from porcine or pig source is one of the non-halal ingredients that commonly found in food and pharmaceutical products. Beside porcine, bovine and fish are also becoming the main commercial sources of gelatin. Since gelatins are derived primarily from porcine and bovine sources, their applications in food and capsules have become issues for some religious adherents such as the Muslim and Jewish communities (Hidaka and Liu, 2003). Non - Islamic countries are the major suppliers and manufacturers of gelatin, with Europe and America being the most prominent manufacturers along with Russia, India, China and Southeast Asia (Cochrane, 2016). In Malaysia, halal food suppliers have to face major challenges in offering their products due to the possible presence of non-halal gelatin (Said et al., 2014). Since the majority of the population in Malaysia are Muslims, the government is fully aware with this fact. As a result, the Department of Islamic Development Malaysia (JAKIM) has launched a Halal Assurance System as a measure in the form of halal certification for each food product (JAKIM, 2015). Nevertheless, it is important to verify the halal status of the ingredients used in the products. Suitable techniques and robust detection methods are

needed for halal authentication purposes (Van der Spiegel et al., 2012). Authentication analysis usually involve analytical techniques which are applicable in dealing with particular samples, depending on the state of the sample itself (Hargin, 1996). Since gelatin is derived from various sources including porcine, bovine and fish, it is paramount to develop a rapid and easy method to distinguish these types of gelatin for halal authentication purposes.

Numerous chemical and chemometrics methods have been applied by researchers in differentiating the sources of gelatins. The most reliable chemical method for gelatin identification is Fourier transform infrared (FTIR) spectroscopy (Hashim et al., 2010). According to Irfanita et al. (2017), the FTIR method is reliable and effective for authentication purposes, due to its ability to offer rapid results and unique spectrum of samples. In addition, FTIR has been widely used in analysis involving gelatin (Premanandh and Bin Salem, 2017). However, chemometrics method is still needed in most chemical analyses, including FTIR, in order to further differentiate the gelatins by analyzing the obtained chemical dataset. The FTIR spectroscopy method produces result in the form of spectrum which contains information of chemical compositions of samples. However, only small differences of FTIR spectra of gelatin can be observed due to the large similarity in their protein structure (Hashim et al., 2010). Therefore, chemometrics analysis using principal component analysis (PCA) is commonly used in combination with FTIR. However, chemometrics using fuzzy graph technique has never been reported in literatures. In this study, a graph representation of FTIR spectra using fuzzy autocatalytic set (FACS) is developed in order to differentiate bovine, porcine and fish gelatins. The implementation of the FACS in FTIR spectra involves the development of its dynamic graph which will be evaluated to identify the dominant features. The identification of the dominant factors and signatures for bovine, porcine and fish gelatin spectra are determined that in turn are deduced for halal authentication purposes.

1.2 Statement of Problem

Identification of gelatins for halal authentication purposes is important due to the concerns on the presence of prohibited ingredients in food and pharmaceutical products. Food such as desserts, jelly, dairy products, and capsule shells in most pharmaceutical products usually contain or made up of gelatin. Furthermore, possible mislabeling of the food's ingredients is an issue itself since most common gelatin source is non-halal porcine that is made up of pig derivative. Moreover, the sources of gelatin possess similar properties and hard to differentiate. Hence, it is important to identify the sources of the gelatin correctly. The FTIR is one of the most reliable chemical methods for gelatin identification and it is often being used with PCA. However, PCA requires pre-processing which is tedious and time consuming. Hence, a new and fast but yet effective chemometrics method is needed for gelatin authentication purposes. Chemometrics statistical methods based are commonly used. However, chemometrics fuzzy graph approach has never been reported in literatures. Thus, a chemometrics technique using fuzzy graph that is faster and rigor than former is introduced in this study.

1.3 Research Objectives

The objectives of the research are:

- (a) To identify bovine, porcine and fish gelatin spectra using Fourier Transform Infrared (FTIR) spectroscopy.
- (b) To construct fuzzy graph representation of the FTIR spectra of the gelatins.
- (c) To develop chemometrics fuzzy autocatalytic set (c-FACS) technique to identify the signature differences between bovine, porcine and fish gelatins.
 - (i) To write an algorithm of the developed chemometrics fuzzy autocatalytic set (c-FACS).
 - (ii) To code the c-FACS algorithm.

- (d) To evaluate and compare the performance of c-FACS for bovine, porcine and fish gelatins identifications against the principal component analysis.

1.4 Scope of Research

The scope of this study is on identification of bovine, porcine, and fish gelatins using FTIR and chemometrics fuzzy autocatalytic set (c-FACS).

1.5 Significance of Research

This study is expected to identify the dominant and signature characteristics of bovine, porcine and fish gelatins which contribute to the authentication of the gelatin sources. The identification of the differences between the gelatin sources is crucial for halal authentication purposes and contributes towards halal science studies. This study is also expected to contribute towards the advancement of fuzzy autocatalytic set (FACS) theory and its applications.

1.6 Research Outline

This thesis consists of six chapters. The first chapter is the introductory chapter which includes the background of research, problem statement, research objectives, scope and significance of research.

Chapter 2 presents the literature review on the current available methods for authentication of gelatin and fuzzy graph. A review on earlier studies involving chemical and chemometrics methods on authentication of gelatin are presented in this chapter. Moreover, theoretical foundation for fuzzy autocatalytic sets (FACS) and transformation to its coordinated form are also discussed in this chapter.

Chapter 3 discusses the experimental procedure and data collection of bovine, porcine and fish gelatins samples using Fourier transform infrared (FTIR) spectroscopy. Principal component analysis (PCA) method is performed on the gelatins spectra and the results will be compared against the proposed method later in Chapter 5.

Chapter 4 is on the development of chemometrics FACS (c-FACS) technique. In this chapter, FTIR spectrum is represented in a form of graph. Each variable and its connectivity in the FTIR system are identified accordingly. Some definitions and theorems regarding the c-FACS matrix are developed and presented in this chapter.

Chapter 5 elaborates on the implementation of the c-FACS. Each gelatin spectrum obtained from FTIR is analyzed using a developed algorithm to identify its dominant and signature features. The classification of the gelatins are presented in Euclidean space. The results and performances of the technique are compared to principal component analysis (PCA).

Finally, the summary and the conclusion of the research are presented in Chapter 6 together with some recommendations. The structure of the thesis is outlined in Figure 1.1.

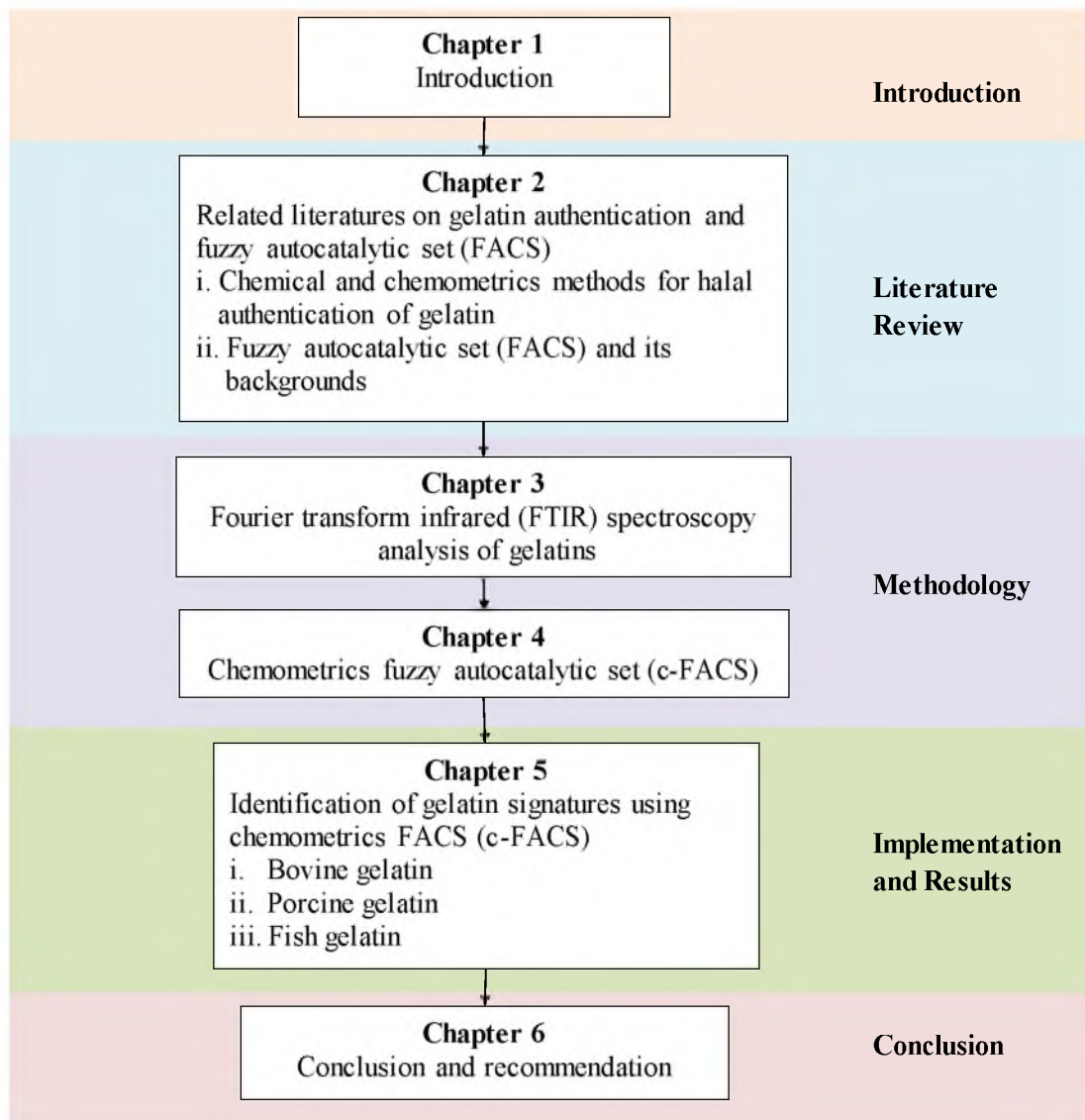


Figure 1.1 Thesis outline

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