UNIVERSITI PUTRA MALAYSIA

CHEMICAL PROFILES, SENSORY ATTRIBUTES AND EFFECT OF MARINATING TIME ON THE FORMATION OF HETEROCYCLIC AMINES OF FOUR TYPES OF SOY SAUCE IN ROASTED CHICKEN

NUR SYIFAA BT ALAM SHAH

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By

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Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfillment of the requirements for the degree of Master of Science

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Chair : Prof. Jinap Selamat, PhD
Faculty : Food Science and Technology

Soy sauce is a fermented condiment used by many Asian countries due to its salty and distinct tasty taste called umami. As soy sauce has been widely used in Southeast Asian cooking, it is important to well-characterize them based on their chemical compositions. Compounds such as free amino acids and glucose which formed during soy sauce fermentation have significant effects on the flavor and taste of soy sauce. However, these compounds also may act as precursor to increase the formation of heterocyclic amines (HCAs). HCAs are potent mutagens that can be easily form in cooked food and a risk factor for human cancers. The aim of this study was to determine the chemical characteristics and sensory attributes of different types of soy sauce (sweet, salty, light and dark) and their effects as marinating ingredient on the formation of HCAs in roasted chicken. The chemical profiling of four different types of soy sauce which are widely consumed and commercially available in Southeast Asia were determined. Sweet, salty, light and dark soy sauce were discriminated based on their chemical characteristics such as sodium chloride, sugars, organic acids, total nitrogen and free amino acids. Sodium chloride was determined by titration, sugar and organic acid by High Performance Liquid Chromatography (HPLC), free amino acids by Gas Chromatography (GC) and total nitrogen by Kjeldhal method. The sensory attribute was studied using the Quantitative Descriptive Analysis (QDA) and the correlation with chemical profiles was analyzed by orthogonal partial least square discriminant analysis (OPLS-DA). In HCAs analyses, chicken breast samples were marinated with sweet, salty, light and dark soy sauce at 0, 3, 6 and 12 hours. The concentration of free amino acids, sugars and creatinine in marinated samples were determined before roasting while HCA's concentration were determined in roasted chicken samples. Unmarinated chicken breasts were treated in similar conditions to serve as reference sample. Result showed that sugars (fructose, glucose and sucrose) were found to be dominant compounds in sweet (35.50 - 64.52 g/100 ml), dark (14.19 - 30.73 g/100 ml) and salty soy sauce (7.27 - 21.74 g/100 ml). The sensory attributes such as color, caramel

odor, viscosity and sweetness taste has increased the overall acceptance in these types of soy sauce. In light soy sauce, sodium chloride (11.69 - 12.58 g/100 ml), total nitrogen (0.52 - 0.88 g/100 ml) and free amino acids (331.20 - 1053.46 mg/100 ml) appeared to be dominant compounds. It was found that saltiness and umami taste were the important sensory attributes that well-characterized the taste of light soy sauce. In HCAs analyses, results showed that all types of soy sauce significantly increased ($p \leq 0.05$) the concentration of HCAs in roasted chicken with increasing marinating time. The highest total concentration of HCAs was found in samples marinated with light soy sauce at 12 hours followed by dark, salty and sweet and the percentage of increment were up to 887%, 375%, 193% and 169% respectively. PhIP showed a complete reduction by 100% in samples marinated with sweet soy sauce at 0 hour. Variations in raw materials used and different production process of different types of soy sauce may contribute to the different chemical profiles and sensory attribute of soy sauce. Free amino acid was found to be strongly correlated with the formation of HCAs than that of reducing sugar and creatinine. The present study suggests that reducing marinating time with soy sauce is important to control the higher formation of HCAs in roasted chicken. Sweet soy sauce could be a better selection of the types of soy sauce to be used as marinating ingredient in the preparation of roasted chicken.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Master Sains

PROFIL KIMIA, SIFAT SENSORI DAN KESAN MASA PEMERAPAN TERHADAP PEMBENTUKAN HETEROSIKLIK AMINES PADA EMPAT JENIS KICAP DI DALAM AYAM PANGGANG

Oleh

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Pengerusi : Prof. Jinap Selamat, PhD
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Kicap soya adalah bahan perasa yang diperam dan digunakan di kebanyakan negara-negara Asia kerana rasa lazat, masin dan rasa yang berbeza dipanggil umami. Kicap soya telah digunakan secara meluas dalam masakan di Asia Tenggara. Oleh itu, ianya adalah penting untuk yang mencirikan mereka berdasarkan komposisi kimia tertentu. Sebatian kimia seperti asid amino bebas dan glukosa yang terbentuk semasa penapaian kicap mempunyai kesan yang besar ke atas rasa kicap soya. Walau bagaimanapun, sebatian kimia ini juga boleh bertindak sebagai prekursor bagi meningkatkan pembentukan heterosiklik amina (HCAs). HCAs adalah mutagen kuat yang mudah terbentuk di dalam makanan yang dimasak dan ianya merupakan faktor risiko terhadap pembentukan kanser pada manusia. HCAs adalah mutagen yang kuat yang boleh dengan mudah terbentuk di dalam makanan yang telah dimasak dan faktor risiko untuk kanser manusia. Tujuan kajian ini adalah untuk menentukan ciri-ciri kimia pada jenis kicap soya yang berbeza (manis, masin, cair dan pekat) dan kesannya sebagai bahan pemerapan dalam pembentukan HCAs di dalam ayam panggang. Profil kimia bagi empat jenis kicap soya yang sering digunakan dan boleh didapati secara komersial di Asia Tenggara telah ditentukan. Kicap soya manis, masin, cair dan pekat telah didiskriminasi berdasarkan ciri-ciri kimia seperti natrium klorida, gula, asid organik, jumlah nitrogen dan asid amino bebas. Natrium klorida ditentukan dengan pentitratan, kepekatan gula dan asid organik telah dianalisa oleh *High Performance Liquid Chromatography tinggi (HPLC),* asid amino bebas oleh *Gas Chromatography (GC)* dan jumlah nitrogen dianalisa melalui kaedah Kjeldhal. Sifat deria dikaji menggunakan *Quantitative Descriptive Analysis (QDA)* dan korelasi dengan profil kimia dianalisa dengan *orthogonal partial least square discriminant analysis (OPLS-DA).* Di dalam analisis HCAs, sampel dada ayam telah diperap dengan menggunakan kicap soya manis, masin, cair dan pekat pada 0, 3, 6 dan 12 jam. Kepekatan amino asid bebas, gula dan kreatinin pada sampel ayam yang diperap telah ditentukan sebelum memanggang manakala kepekatan HCA pula ditentukan di dalam sampel ayam panggang. Dada ayam yang tidak diperap

disediakan dalam keadaan yang sama seperti sampel kajian untuk digunakan sebagai sampel rujukan. Keputusan menunjukkan bahawa gula (fruktosa, glukosa dan sukrosa) merupakan sebatian kimia yang dominan di dalam kicap soya manis (35.50 - 64.52 g/100 ml), diikuti dengan kicap soya pekat (14.19 - 30.73 g/100 ml) dan kicap soya masin (7.27 - 21.74 g/100 ml). Nilai rasa seperti warna, bau karamel, kelikatan dan rasa kemanisan telah meningkat penerimaan keseluruhan bagi semua jenis kicap soya. Dalam kicap soya cair, natrium klorida (11.69 - 12.58 g/100 ml), jumlah nitrogen (0.52 - 0.88 g/100 ml) dan asid amino bebas (331.20 - 1053.46 mg/100 ml) merupakan sebatian dominan. Didapati bahawa kemasinan dan rasa umami adalah nilai rasa penting dalam mengkarakterisasi rasa kicap soya cair. Di dalam HCAs analisis, hasil kajian menunjukkan bahawa kepekatan HCAs meningkat dengan ketara ($p \leq 0.05$) dengan peningkatkan masa pemerapan pada semua jenis kicap soya yang digunakan untuk memerap ayam panggang. Jumlah kepekatan tertinggi HCAs ditemui pada sampel diperap dengan kicap soya cair pada tempoh 12 jam diikuti dengan kicap pekat, masin dan manis dan peratusan kenaikan meningkat, masing-masing kepada 887%, 375%, 193% dan 169%. PhIP menunjukkan pengurangan lengkap sebanyak 100% di dalam sampel yang diperap dengan kicap manis pada tempoh 0 jam. Variasi dalam bahan-bahan mentah yang digunakan dan proses pembuatan yang berbeza untuk jenis kicap soya yang berbeza boleh menyumbang kepada profil kimia dan sifat deria kicap yang berlainan. Asid amino bebas telah didapati mempunyai kaitan yang sangat rapat terhadap pembentukan HCAs berbanding dengan gula dan kreatinin. Kajian ini menunjukkan bahawa pengurangan masa pemerapan dengan kicap soya adalah penting untuk mengawal pembentukan dan kepekatan HCAs yang lebih tinggi di dalam ayam panggang. Kicap soya manis didapati merupakan pilihan yang lebih baik berbanding jenis kicap soya yang lain untuk digunakan sebagai bahan pemerapan dalam penyediaan ayam panggang.

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I certify that a Thesis Examination Committee has met on 05 May 2016 to conduct the final examination of Nur Syifaa binti Alam Shah on her thesis entitled "Chemical Profiles, Sensory Attributes and Effect of Marinating Time on the Formation of Heterocyclic Amines of Four Types of Soy Sauce in Roasted Chicken" in accordance with the Universities and University Colleges Act 1971 and the Constitution of the Universiti Putra Malaysia [P.U.(A) 106] 15 March 1998. The Committee recommends that the student be awarded the Master of Science.

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

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Signature: _____
Name of Member of Supervisory Committee: Dr. Lasekan Olusegun

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LIST OF ABBREVIATIONS

%	Percentage
°C	Degree celcius
µg	microgram
µl	microlitre
µm	micrometer
ANOVA	Analysis of variance
AOAC	Association of Official Analytical Chemist
AαC	2-amino-9H-pyrido [2,3- b]indole
cm^3	Cubic centimetres
DiMeIQx	2-Amino-3,4,8-trimethyl-3H-imidazo[4,5-f]quinoxaline
DNA	Deoxyribonucleic Acid
ESI+	Positive electrospray ionisation
FAO	The *Food and Agriculture Organization* of the United Nations
g	Gram
Glu-P-1	2-amino-6-methyldipyrido-[1,2-a:3',2'-d]imidazole
Glu-P-2	2-aminodipyrido-[1,2-a:3',2'-d] imidazole
Harman	1-methyl-9H-pyrido [4,3-b] indole
HCAs	Heterocyclic Amines
HPLC	High-performance liquid chromatography
HSD	Honest significant difference
IARC	The International Agency for Research on Cancer
IQ	2-amino-3-methylimidazo[4,5-f] quinoline
IQx	2-amino-3-methylimidazo[4,5-f]-quinoxaline
ISO	The International Organization for Standard
kg	kilogram
LC-MS/MS	Liquid Chromatography tandem Mass Spectrometer
LOD	Limit of detection
max	maximum
MeIQ	2-amino-3-4-di-methyl-imidazo[4,5-f]quinoline
MeIQx	2-amino-3,8-dimethyl-imidazo[4,5-f]quinoxaline
mg	miligram
min	minimum
ml	mililitre
mM	milimole
MRM	Multiple reaction monitoring
MS	Malaysian Standard
MSG	Monosodium glutamate
ng	Nanogram
nmol	Nanomole
Norharman	9H-pyrido[4,3- b]indole
NTP	The National Toxicology Program

OPLS-DA	Orthogonal partial least square discriminant analysis
PhIP	2-Amino-1-methyl-6-phenylimidazo[4,5-b]pyridine
PLS	Partial least square regression
ppb	Part per billion
ppm	Part per million
QDA	Quantitative descriptive analysis
RI	Refractive index
rpm	Revolutions per minute
TEA	Triethylamine
Trp-P-1	3-amino-1,4-dimethyl-5H-pyrido[4,3-b]- indole
Trp-P-2	3-amino-1-methyl-5H-pyrido[4,3-b]indole
UHPLC	Ultra High Performance Liquid Chromatography
USDA	The United States Department of Agriculture
UV	Ultraviolet
v/v	Volume per volume
w/v	Weight per volume
WHO	The World Health Organization

CHAPTER 1

INTRODUCTION

1.1 Background of Study

Soy sauce is a traditional condiment produced by fermenting soybeans along with roasted grain, water and salt. It has been integrated into the traditional cuisines of many Asian cultures and in more recent times it is used in western cuisine and prepared foods. There are many varieties of soy sauce produced around the world. In Southeast Asia the most typical types of soy sauce used are the Indonesian-type; sweet and salty (also known as '*kecap manis'* and *'kecap asin'*) and the Chinese-type; light and dark soy sauce (Shurtleff & Aoyagi, 2012). In addition, the quality of each types of soy sauce differs in terms of their chemical compositions due to the different of raw material used and production method (Feng et al., 2013). This study provides a comprehensive chemical profiling and sensory attributes of four different types of soy sauce namely sweet, salty, light and dark type. Soy sauce also is frequently used in marinating, stir-fried cooking and as at table condiment due to its salty and distinct taste called umami (Lioe et al., 2004). As soy sauce has many functions in cooking, this study also discussed the effect of soy sauce as marinating ingredient in the formation of heterocyclic amines (HCAs) in roasted chicken. HCAs are carcinogenic compounds that are usually formed at ppb level in numerous cooked foods commonly consumed in the diet. They present in protein-rich foods especially from animal-based when the cooking temperature exceeds 150°C (Skog, 1998). The formation of HCAs occurs via Maillard reaction involving precursors such as creatinine, free amino acids and sugars (Murkovic, 2004). Up to date, more than 25 HCAs have been identified in different cooked muscle foods and the most common HCAs found are MeIQ (2-amino-3-4-di-methyl-imidazo[4,5-f]quinoline), MeIQx (2-amino-3,8-dimethyl-imidazo[4,5-f]quinoxaline), PhIP (2-Amino-1-methyl-6-phenylimidazo[4,5-b]pyridine) and AαC (2-amino-9H-pyrido [2,3- b]indole) (Knize et al., 1994). These HCAs have been listed as possible human carcinogens (class 2B) and IQ (2-amino-3-methylimidazo[4,5-f] quinoline) as a probable human carcinogen (class 2A) by The International Agency for Research on Cancer (IARC, 1993).

1.2 Problem Statements

Sweet, salty, light and dark types of soy sauces are very popular and commonly used in Southeast Asian cooking. However, there is limited information reported in literature on these types of soy sauces. The chemical characteristics, sensory attributes and production method of these four types of soy sauce also have not been clarified thoroughly. Although there were several studies conducted on the chemical characteristics of soy sauce, most literatures focusing on Japanese-types of soy sauce only such as *koikuchi, usukuchi, tamari and shiro shoyu* (Luh, 1995). In addition, the quality standard for soy sauce has not been established extensively in other country except Japan. In Japan, soy sauce were classified into three quality standards such as

'Special grade', 'Upper grade' and 'Standard grade' depending on the amount of wheat and soybean used as the raw materials (Luh, 1995). The classification of soy sauce in Southeast Asia is usually made according to the convenience of manufacturers only (Judoamidjojo et al., 1985). Although there are specifications standard issued in The Malaysian Standard MS 2042, this standard prescribed the specification for salty soy sauce only whereas sweet, light and dark soy sauce are commonly used by Malaysian. To the best of our knowledge, this study also discussed on the safety aspect of soy sauce as marinating ingredient in roasted chicken. It has been reported that the fermentation products of soy sauce like free amino acids and reducing sugars possessed an initiating effect on the formation of HCAs in cooked foods (Lan & Chen, 2002). Soy sauce is one of the popular ingredients that usually used to marinate roasted chicken. However, there is limited information reported on the effect of soy sauce on the formation of HCAs. Furthermore, the type of soy sauce used also have not been mentioned clearly in any literatures whereas there are many varieties of soy sauce that are commonly used in cooking such as sweet, salty, light and dark type.

1.3 Significant of Study

The content of total nitrogen, free amino acid, organic acid, salt and reducing sugar are the important parameters that can be used to determine the quality and flavour characteristics of soy sauce (Liu, Bao, & Ren, 2011). Therefore, in this study an attempt was made to provide more data which will serve to the standardization and quality improvement in the production of different types of soy sauce. As soy sauce has high concentration of free amino acid, it is extremely important to study its effect on the formation of HCAs in roasted chicken. The findings in this study should be of interest to the soy sauce manufacturer, food processing industry, restaurateurs, and home-makers as it can be used as general precaution in using soy sauce as marinating ingredient in the preparation of roasted chicken.

1.4 Objectives

Therefore, the objectives of this study were:

1. To determine the chemical characteristics and sensory attributes of sweet, salty, light and dark types of soy sauce and discriminate the differences between them.
2. To evaluate the chemical characteristics of different types of soy sauce and the relationship with its sensory attributes.
3. To determine the effect of different types of soy sauce and marinating times on the formation of HCAs in roasted chicken.

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