



**UNIVERSITI PUTRA MALAYSIA**

**EFFECTS OF EDIBLE OIL AND FAT-SOLUBLE VITAMINS ON  
ACRYLAMIDE FORMATION IN A CHEMICAL MODEL SYSTEM AND  
DURING INTERMITTENT DEEP-FAT FRYING OF FRENCH FRIES**

**KUEK SU LEE**

**FSTM 2020 14**



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By

**KUEK SU LEE**

**Thesis submitted to the School of Graduate Studies, Universiti Putra Malaysia, in  
Fulfillment of the Requirements for the Degree of Master of Science**

**January 2020**

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

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**January 2020**

**Chairperson: Maimunah Sanny, PhD**  
**Faculty : Food Science and Technology**

High concentration of acrylamide had been identified in fried potato products. However, the effect of edible oils and fat-soluble vitamins toward acrylamide had yet to be determined clearly. The objectives of this study were firstly to determine the contributions of edible oils in terms of type of vegetable oils and number of frying cycles toward acrylamide formation. Second objective is to determine the relationship of fatty acid composition (FAC) and total polar compound fraction (TPC) of vegetable oils on acrylamide during intermittent deep-fat frying of French fries. The third objective was to determine effect of different levels of vitamin A and E homologs on acrylamide formation in Asparagine-Glucose model system. Par-fried potato strips were fried in four types of vegetable oils i.e., palm olein, red palm olein, sunflower oil and soybean oil for 80 cycles. Acrylamide concentration in French fries were analysed. Oils used every 16<sup>th</sup> cycle of frying were analysed for peroxide value (PV), *p*-anisidine value (*p*-AV), total oxidation value (TOTOX) and free fatty acid (FFA). Consequently, frying cycle significantly affected acrylamide concentration in all oils except palm olein. A weak positive, however significant correlation were found between acrylamide with *p*-AV and FFA; but not PV and TOTOX. Furthermore, FFA contribute more to acrylamide formation compare to *p*-AV. For the second study, FAC and TPC of vegetable oils at every 16<sup>th</sup> frying cycle were analysed. Saturated fatty acid (SFA) increased and polyunsaturated fatty acid (PUFA) decreased as number of frying cycle increased for all oils. Significant positive correlation was observed between SFA, monounsaturated fatty acid (MUFA), PUFA and acrylamide concentration with PUFA being the strongest. TPC and polar compound fractions in terms of polymerised triacylglycerol (PTAG), oxidised triacylglycerol and diacylglycerol of all vegetable oils increased significantly across 80 frying cycles. Weak but significant negative correlation was observed between PTAG of vegetable oils and acrylamide. For the third study, six vitamin homologs, i.e., beta-carotene (BC), alpha-tocopherol (AT), delta-tocopherol (DT), alpha-tocotrienol (ATT), delta-tocotrienol (DTT) and gamma-tocotrienol (GTT), were individually added at the level of 1 and 10 µmol into the model system and heated at 160°C for 20 minutes. AT, DT and GTT at 1 µmol significantly increased acrylamide concentration when compared to control with AT being the highest. All homologs at 10 µmol except DT significantly

increased acrylamide with BC being the highest. BC significantly increased acrylamide when its level increased. AT, DT and GTT significantly decreased acrylamide when their levels increased. Vitamin A and E homologs enhanced acrylamide formation. However, its levels can determine their effects as antioxidant or pro-oxidant. Thus, edible oils during frying as well as fat-soluble vitamins play a significant role in acrylamide formation.



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

**KESAN-KESAN MINYAK DAN VITAMIN LARUT MINYAK DI DALAM  
MINYAK SAYURAN DALAM PEMBENTUKAN AKRILAMIDA DI DALAM  
SISTEM MODEL KIMIA DAN KENTANG GORENG**

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Kandungan akrilamida yang tinggi telah dikenalpasti dalam produk kentang goreng. Walau bagaimanapun, kesan minyak masak dan vitamin larut lemak di dalamnya terhadap akrilamida belum ditentukan dengan jelas. Objektif pertama kajian ini adalah untuk menentukan sumbangan lipid terhadap pembentukan akrilamida dalam proses penggorengan kentang. Objektif yang kedua adalah untuk menentukan hubungan antara komposisi asid lemak (FAC) dan jumlah pecahan sebatian kutub (TPC) dalam minyak sayuran terhadap pembentukan akrilamida dalam proses penggorengan kentang. Objektif ketiga kajian ini adalah untuk menentukan kesan pelbagai tahap vitamin A dan E homolog terhadap pembentukan akrilamida dalam sistem model Asparagine-Glucose. Kentang digoreng dengan menggunakan empat jenis minyak sayuran iaitu minyak sawit, minyak bunga matahari, minyak sawit merah dan minyak kacang soya selama 80 penggorengan. Kandungan akrilamida telah diukur. Nilai peroxida (PV), nilai *p*-anisidin (*p*-AV), nilai pengoksidan total (TOTOX) dan asid lemak bebas (FFA) untuk minyak yang telah digunakan setiap 16 penggorengan telah diukur. Akibatnya, tahap pusingan penggorengan boleh mempengaruhi pembentukan akrilamida dalam semua minyak kecuali minyak sawit. Korelasi dan sumbangan positif yang lemah, didapati antara acrylamide dengan *p*-AV dan FFA; tetapi tidak kepada PV dan TOTOX. FFA menyumbang lebih banyak kepada pembentukan akrilamida berbanding dengan *p*-AV. Untuk kajian kedua, asid lemak tepu (SFA) meningkat dan asid lemak tidak jenuh (PUFA) berkurangan apabila bilangan kitaran goreng meningkat. Hubungan positif yang signifikan didapati antara SFA, asid lemak tidak jenuh tunggal (MUFA) dan PUFA minyak sayuran dan akrilamida. Korelasi PUFA juga paling tinggi berbanding dengan SFA dan MUFA. TPC untuk semua minyak sayuran meningkat selama 80 kitaran goreng. Ujian triacylglycerol polimer (PTAG), triacylglycerol teroksida (OTAG) dan diacylglycerol (DAG) meningkat dengan bilangan penggorengan. Korelasi negative diperhatikan antara PTAG minyak sayuran dan akrilamida. Untuk kajian ketiga, enam vitamin homolog, iaitu beta-karoten (BC), alpha-tocopherol (AT), delta-tokoferol (DT), alpha-tocotrienol (ATT), delta-tocotrienol (DTT), dan gamma-tocotrienol (GTT) ditambah secara individu pada tahap 1 dan 10  $\mu\text{mol}$  ke dalam sistem model dan dipanaskan pada 160 °C selama 20 minit. AT, DT dan GTT pada 1  $\mu\text{mol}$  meningkatkan

akrilamida apabila dibandingkan dengan AT yang tertinggi. Semua homolog pada 10  $\mu\text{mol}$  kecuali DT meningkatkan akrilamida dengan BC yang tertinggi. BC meningkat dengan ketara akrilamida apabila tahapnya bertambah. AT, DT dan GTT menurun dengan ketara akrilamida apabila tahap mereka meningkat. Vitamin A dan E homolog meningkatkan pembentukan akrilamida. Walau bagaimanapun, parasnya boleh menentukan kesannya sebagai antioksidan atau pro-oksidan. Lipid dan vitamin larut lemak dalam minyak mempengaruhi pembentukan akrilamida.



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I certify that a Thesis Examination Committee has met to conduct the final examination of Kuek Su Lee on her thesis entitled “Effect of lipid and fat-soluble antioxidant in vegetable oils on acrylamide formation in a chemical model system and during intermittent deep-fat frying of French fries” 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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## LIST OF ABBREVIATIONS

°C	Degree celcius
µg	Microgram
µl	Microliter
µmol	Micromole
%	Percentage
APA	Aminopropionamide
ANOVA	Analysis of variance
AOCS	American Oil Chemists' Society
ASTM	American Society for Testing and Materials
BHA	Butylated hydroxylanisole
BHT	Butylated hydroxytoluene
cm	Centimeter
DAG	Diacylglycerol
DNA	Deoxyribonucleic acid
EGCG	Epigallocatechin gallate
EFSA	European Food Safety Authority
FAC	Fatty acid composition
FAME	Fatty acid methylester
FAO	Food and Agriculture Organisation
FFA	Free fatty acid
g	Gram
GC	Gas Chromatography
HLB	Hydrophilic-Lipophilic-Balanced
HPLC	High Performance Liquid Chromatography
IARC	International Agency for Research on Cancer
IUPAC	International Union for Pure and Applied Chemistry
kg	Kilogram
kPa	Kilopascal

M	Meter
MCX	Mixed-mode Cation-eXchange
mg	Milligram
ml	Milliliter
mm	Millimeter
mmol	millimole
MUFA	Monounsaturated fatty acid
<i>m/z</i>	Mass to charge ratio
ng	Nanogram
OTAG	Oxidised triacylglycerol
<i>p</i> -AV	<i>p</i> -anisidine value
PTAG	Polymerised triacylglycerol
PUFA	Polyunsaturated acid
PV	Peroxide value
RCF	Relative centrifugal force
RPO	Red palm oil
SAPP	Sodium acid pyrophosphate
SFA	Saturated fatty acid
SNFA	Swedish National Food Authority
TAG	Triacylglycerol
TBHQ	Tert-butyl hydroquinone
TOTOX	Total oxidation value
TPC	Total polar compounds
W	Heat transfer rate
WHO	World Health Organisation

# CHAPTER 1

## INTRODUCTION

### 1.1 Background of study

Acrylamide has been considered as potential carcinogenic substance to human as it can significantly causes cancer in experimental animals (IARC, 1994). Exposure of humans to acrylamide can originate from ingesting food or drink, through dermal contact with acrylamide-containing materials, and by inhaling acrylamide vapors or particles (Manson et al., 2005). Acrylamide can be formed with presence of its precursor that are free asparagine and reducing sugars such as glucose and fructose under heated condition. The process is known as Maillard reaction (Friedman, 2016). Lipid oxidation also serve as another pathway for acrylamide formation in foods (Gert and Klostermann, 2002). Acrylamide is predominantly found in food high in carbohydrate that had undergo high temperature treatment (Tarake et al., 2002). Potato-rich foods that are baked or fried such as French fries observed to contain high acrylamide concentration and can be considered as the most common source of foods that contribute to acrylamide (Biedermann et al., 2002). Frying is the main method for the preparation of French fries and is one of the universal cooking technique in food preparation (Ahmad Tarmizi et al., 2006). Intermittent frying generally refers to performing prolong frying of foods at predetermined time interval where the oil is heated and kept after use for future frying (Seppanen and Csallany, 2006). However, prolong frying process that occurs under the presence of oxygen, moisture, trace elements and free radical can cause deterioration of oils quality (Andrikopoulos et al., 2002).

### 1.2 Problem statement

Since fried food absorbs considerable amount of oil, quality loss with regard to frying oil would not only exerts undesired effects on its sensory attributes (Gertz & Behmer, 2014) but also elicits unwanted breakdown derivatives such as acrylamide that can adversely affect consumer health (Zribi et al., 2014). Lipid oxidation products with the presence of carbonyl groups, such as aldehydes and ketones, could form acrylamide with asparagine despite absence of reducing sugars (Ehling & Shibamoto, 2005; Yasuhara, Tanaka, Hengel & Shibamoto, 2003).

Different types of vegetable oils used for frying also affect acrylamide formation through the increase or decrease in its concentration (Gert and Klostermann, 2002). For instance, olive oil was shown to contribute to higher acrylamide content compared to corn oil during frying of potato chips (Becalski et al, 2003). Gertz & Klostermann (2002) also reported that use of palm oil increased acrylamide concentration compared to rapeseed and sunflower oil during frying of French fries. Furthermore, some researchers also reported that number of frying cycle performed during frying process could affect the formation of acrylamide in French fries (Gertz, 2004) although some researchers disagree on this statement (William, 2005). Thus, effects of frying components such as

types of vegetable oil used for frying, number of frying cycle and lipid deterioration process in terms of oxidation and hydrolysis have yet to be elucidated clearly in any context. Composition of fatty acids in vegetable oils towards formation of acrylamide had also not being evaluated clearly.

Besides that, vegetable oils have been reported to be rich in fat-soluble vitamin such as vitamin A and E (Kalyana et al., 2003). Researchers have reported that antioxidants could inhibit lipid oxidation, reduce formation of carbonyls and thus limit acrylamide formation (Kocadağlı et al., 2012). Some examples demonstrated were such as the use of extract of bamboo leaves (Zhang and Zhang, 2008), ascorbic acid (Biedermann et al., 2002), and rosemary oil (Hedegaard et al., 2008) can effectively reduce formation of acrylamide. Discordant effects of antioxidants towards acrylamide were also being reported. For instance, addition of butylated hydroxytoluene (BHT) and sesamol into meat before heat treatment increase acrylamide concentration (Tareke, 2003). However, the effect of vitamin A and E especially in terms of their homolog forms towards acrylamide has yet to be determined by researcher. Therefore, the effect of all components that constitute the frying process on acrylamide formation will be clarified in this study.

### **1.3 Significant of study**

Through this study, research gap regarding the effect of lipid towards acrylamide formation during frying can be filled. Besides that, effects of vitamin A and E toward acrylamide can also be understand clearly through the use of model system. As acrylamide had been listed by International Agency for Research on Cancer (IARC) as Group 2A carcinogen (IARC, 1994), understanding on its formation during frying can improve the safety attribute of the food as well as the frying process. The findings of this study can undoubtedly contribute the public health as frying is widely used as food preparation method for most of the domestic household, food service establishment and also industry.

### **1.4 Objectives**

Therefore, the objectives of this study were:

- i. To identify the contribution of edible oils in terms of its type and number of frying cycle towards acrylamide formation during intermittent deep-fat frying of French fries.
- ii. To evaluate the relationship between fatty acid composition and total polar compound fraction of vegetable oils on acrylamide formation during intermittent deep-fat frying of French fries.
- iii. To evaluate the effect of vitamin A and E homologs at different levels on acrylamide in a chemical model system.

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Kuek Su Lee was born in Muar, Johor, Malaysia on 23 September 1992. She received her primary education at SJK(C) Chung Hwa 2(B), Muar, Johor. She then continued her five years secondary education at SMK Convent Muar, Johor. After secondary school, she attended pre-university education at SMK Dato Sri Amar Diraja, Muar, Johor. She began her higher education at Universiti Putra Malaysia and acquired Bachelor Degree in Food Science and Technology in year 2016. After graduation, she worked as QA executive at Tatawa Industries (M) Sdn. Bhd. and Pacific Food Products Sdn. Bhd., respectively. She then continued to pursue her Master Degree in Food Safety at Universiti Putra Malaysia.



## LIST OF PUBLICATION

### Publication

Kuek S.L., Ahmad Tarmizi A.H., Abd Razak R.A., Jianp S., Saporin N., Sanny M. (2020). Contribution of lipid towards acrylamide formation during intermittent deep-fat frying of French fries. *Food Control*, 118, 107430, 1 – 8.

### Submitted for publication

Kuek S.L., Ahmad Tarmizi A.H., Abd Razak R.A., Jianp S., Saporin N., Sanny M. Relationship between fatty acid composition and total polar compounds fraction of vegetable oils on acrylamide formation during intermittent deep-fat frying of French fries. Preparation for submission to *Food Chemistry*.

Kuek S.L., Ahmad Tarmizi A.H., Abd Razak R.A., Jianp S., Leong WH., Bryan K.S.L., Sanny M. Effect of vitamin A and vitamin E on acrylamide formation in Asparagine-Glucose model system Preparation for submission to *Food Control*.

### Poster presentation

Kuek S.L., Ahmad Tarmizi A.H., Abd Razak R.A., Jianp S., Sanny M. The effect of fat soluble vitamins in vegetable oils on acrylamide during intermittent deep-fat frying of French fries and in a chemical model system. In Food Safety Postgraduate Mobility Program, Bogor, Indonesia, November 2018.

Kuek S.L., Ahmad Tarmizi A.H., Abd Razak R.A., Jianp S., Saporin N., Sanny M. Contribution of lipid towards acrylamide formation during intermittent deep-fat frying of French fries. In MPOB International Palm Oil Congress and Exhibition (PIPOC) 2019, Kuala Lumpur, Malaysia, November 2019.