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DEVELOPMENT OF CHITOSAN-GRAPHENE OXIDE NANOCOMPOSITE FILMS FOR ACTIVE MARGARINE PACKAGING

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By

FOONG HAN LYN

Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in the Fulfilment of the Requirements for the Degree of Doctor of Philosophy

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DEVELOPMENT OF CHITOSAN-GRAPHENE OXIDE NANOCOMPOSITE FILMS FOR ACTIVE MARGARINE PACKAGING

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FOONG HAN LYN

October 2021

Chair : Nur Hanani Zainal Abedin, PhD
Faculty : Food Science and Technology

Chitosan (CS) has gained significant attention as a food packaging material due to its film-forming ability, biocompatibility, and biodegradability. However, its applications have been limited by its weak mechanical properties and hydrophilicity. The aim of this study was to develop a chitosan-graphene oxide (CSGO) nanocomposite film with improved mechanical properties as well as water vapour, oxygen, and light barrier properties in comparison to pure CS film, for the antioxidant active packaging of palm olein-based margarine. In the first objective, GO samples with four different degrees of oxidation were synthesized by controlling the ratio of graphite to the oxidizing agent, potassium permanganate ($KMnO_4$). The sample GO4, synthesized with a 1:8 w/w graphite:$KMnO_4$ ratio was embedded with abundant oxygen-containing groups, as supported by the Fourier-transform infrared (FTIR) and Raman spectra. The addition of GO4 into CS increased ($p < 0.05$) the mechanical strength and UV light barrier of the CS/GO4 composite. In the second objective, the effects of sonication time of GO4 (30, 60, and 120 min) and heating temperature of the films (30, 60, and 120 ºC) on the structural and physical properties of the CSGO4 composites were investigated. After 120 min of sonication in a sonicator bath, graphene oxide nanosheets (GO120) of ~1 nm thick were obtained, as demonstrated using dynamic light scattering (DLS) technique and atomic force microscopy (AFM). The incorporation of GO120 decreased ($p < 0.05$) the light transmittance of CS films whereas heating the composites at 120 °C lowered ($p < 0.05$) the water solubility and water vapour permeability (WVP). All of the films were completely decomposed within 28 days in a soil burial test. In the third objective, trisodium citrate (CIT) and sodium tripolyphosphate (TPP) solutions of different concentrations (0.5, 1.0. 2.0, and 3.0% w/v) were used as crosslinking agents for the films. Successful crosslinking was confirmed by FTIR spectroscopy. The hydrophilicity and light transmittance decreased ($p < 0.05$) with the increase in CIT and TPP. At 3.0% w/v, the elongation at break and tensile strength of the TPP-crosslinked CSGO films increased ($p < 0.05$) by 42 and 82%, respectively, outperforming CIT as a crosslinking agent. In the final objective, the effect of the concentrations of CS (1.5 and 2.0% w/v) and GO4 (0.5, 1.0, and 2.0% w/w CS) on the properties of nanocomposite films were investigated. The WVP and oxygen permeability (OP) decreased ($p < 0.05$) by 43 and 54%, respectively. The antioxidant properties of the composite film increased ($p < 0.05$) with the concentration of GO4, as supported by the DPPH radical scavenging assay. The changes in the peroxide value (PV) and thiobarbituric acid reactive substances (TBARS) of the margarine samples were monitored for 30 d at 4 ºC. For the margarine sample that was wrapped with the GOCS1.5 GO2.0 film (CS 1.5% w/v, GO 2.0% w/w CS), the PV and TBARS values were 36 and 79% lower ($p < 0.05$) in comparison to the low-density polyethylene films. The combination of these properties such as low WVP, OP, and light transmittance, as well as the radical scavenging activities suggests that the CS1.5 GO2.0 film could be a potential antioxidant active packaging for margarine.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk Ijazah Doktor Falsafah

PENBANGUNAN FILEM NANOKOMPOSIT KITOSAN-OKSIDA GRAFENA SEBAGAI PEMBUNGKUSAN AKTIF MAJERIN

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Kitosan (CS) telah mendapat perhatian sebagai bahan pembungkusan makanan kerana keupayaan pembentukan filem, keserasian bio, dan terbiodegradasinya. Walau bagaimanapun, aplikasi kitosan adalah terhad disebabkan oleh sifat mekanikal yang kurang memuaskan dan sifat hidrofiliknya. Projek ini bertujuan untuk membangunkan filem komposit kitosan-dioksida grafena (CSGO) dengan penambahbaikan sifat mekanikal serta pengurangan penembusan wap air, oksigen, dan cahaya, untuk dijadikan pembungkus aktif marjerin. Dalam objektif pertama, sampel-sampel GO yang mempunyai empat tahap pengoksidaan yang berbeza telah dihasilkan dengan penggunaan nisbah-nisbah yang berbeza untuk grafit dengan agen pengoksidaan, iaitu permanganat kalium ($KMnO_4$). Spektroskopi inframerah transformasi *Fourier* (FTIR) dan spektroskopi Raman telah menunjukkan bahawa sampel GO4 yang dihasilkan dengan nisbah 1:8 w/w untuk grafit dan KMnO4 adalah kaya dengan kumpulan-kumpulan berfungsi yang mengandungi oksigen. Penambahan GO4 ke dalam CS telah meningkatkan ($p < 0.05$) kekuatan mekanikal dan mengurangkan ($p < 0.05$) kebolehan penembusan cahaya untuk filem CS. Untuk objektif kedua, kesan-kesan tempoh sonikasi (30, 60, dan 120 min) dan suhu pemanasan (30, 60, dan 120 ºC) terhadap filem komposit telah dikaji. Ujian penyerakan cahaya dinamik (DLS) dan mikroskopi daya atom (AFM) menunjukkan bahawa kepingan nano oksida grafena dengan ketebalan ~1 nm (GO120) berjaya diperolehi selepas sonikasi selama 120 min dalam mandi ultrasonik. Penambahan GO120 mengurangkan ($p < 0.05$) kebolehan penembusan cahaya CS manakala pemanasan filem CSGO pada suhu 120 ºC telah mengurangkan ($p < 0.05$) kelarutan dan kebolehtelapan wap air (WVP). Semua filem komposit terurai sepenuhnya di dalam tanah kompos dalam 28 hari. Dalam objektif ketiga, larutan trinatrium sitrat (CIT) dan natrium tripolifosfat (TPP) dengan kepekatan yang berbeza (0.5, 1.0. 2.0, dan 3.0% w/v) telah digunakan sebagai agen pautan silang untuk filem CS dan CSGO. Kejadian pautan silang telah disahkan melalui spektroskopi FTIR. Sifat hidrofilik dan kebolehan penembusan cahaya menurun ($p < 0.05$) dengan kenaikan kepekatan CIT dan TPP. Untuk filem CSGO yang dirawat dengan TPP, peratusan pemanjangan pada takat putus dan kekuatan mekanikal meningkat ($p < 0.05$) sebanyak 42 dan 82%, menunjukkan bahawa TPP adalah agen pautan silang yang lebih sesuai berbanding dengan CIT. Untuk

objektif yang terakhir, kesan-kesan kepekatan CS (1.5 dan 2.0% w/v) dan GO4 (0.5, 1.0, dan 2.0% w/w CS) telah dikaji. Untuk filem komposit, WVP dan kebolehtelapan oksigen (OP) menurun ($p < 0.05$) sebanyak 43 dan 54%. Aktiviti antioksida meningkat ($p < 0.05$) dengan kepekatan GO4 dan disokong oleh analisis aktiviti memerangkap radikal DPPH. Perubahan nilai peroksida (PV) dan uji asid tiobarbiturik (TBARS) untuk sampel marjerin dipantau selama 30 hari pada 4 ºC. Sampel marjerin yang dibalut dengan filem CS1.5 GO2.0 menunjukkan penurunan ($p < 0.05$) PV dan TBARS sebanyak 36 dan 79% berbanding dengan filem polietilena berketumpatan rendah. Gabungan sifat-sifat filem CSGO seperti WVP, OP, dan penembusan cahaya yang rendah serta aktiviti memerangkap radikal DPPH membuktikan bahawa filem tersebut berpotensi untuk dijadikan pembungkusan aktif antioksida untuk marjerin.

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

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LIST OF ABBREVATIONS

ABTS	2,2'-azino-bis(3-ethylbenzothiazoline-6-sulfonic acid)
AOAC	Association of Analytical Communities
AOCS	American Oil Chemists' Society
APS	Average particle size
ASTM	American Society for Testing and Materials
CA	Crosslinking agent
CIE	International Commission on Illumination
CIT	Trisodium citrate
CMC	Carboxymethyl cellulose
CS	Chitosan
CSGO	Chitosan-graphene oxide
DDA	Degree of deacetylation
DPPH	2,2-diphenyl-1-picrylhydrazyl
DSC	Differential scanning calorimeter
EAB	Elongation at break
EPA	Environmental Protection Agency
FAO	Food and Agriculture Organization
FDA	Food and Drug Administration
GBM	Graphene-based material
GNP	Graphene nanoplatelets
GO	Graphene oxide
GRAS	Generally Recognised as Safe
HDPE	High-density polyethylene
LBL	Layer-by-layer
LDPE	Low-density polyethylene
LLDPE	Linear low-density polyethylene
LOD	Limit of detection
MAP	Modified atmospheric packaging

MDA	Malondialdehyde
MMT	Montmorillonite
OP	Oxygen permeability
PBAT	Polybutylene adipate terephthalate
PE	Polyethylene
PEC	Polyelectrolyte complexes
PEI	Polyethyleneimine
PET	Polyethylene terephthalate
PI	Polyimide
PLA	Polylactic acid
PMMA	Polymethyl methacrylate
PP	Polypropylene
PV	Peroxide value
PVA	Polyvinyl alcohol
rGO	Reduced graphene oxide
RH	Relative humidity
ROS	Reactive oxygen species
SEM	Scanning electron microscopy
SPC	Sustainable Packaging Coalition
TBA	Thiobarbituric acid
TBARS	Thiobarbituric acid reactive substances
TPP	Sodium tripolyphosphate
TS	Tensile strength
UV	Ultraviolet
WVP	Water vapour permeability
XPS	X-ray photoelectron spectroscopy
YM	Young's modulus

CHAPTER 1

INTRODUCTION

1.1 Background

Packaging is an essential part of the food system. It provides containment and protection to the contents against biological, chemical, and physical contaminations throughout the manufacturing and shipping processes, until they reach the final consumer (Robertson, 2010). Without packaging, the process of materials handling would be very inefficient, messy and costly. In addition, it would be almost impossible to convey information of a product to the respective consumers using modern marketing strategies.

With the vast development of technologies, food packaging has evolved from simply a container to hold food to something that can play an active role in food quality (Figure 1.1). Active packaging improves the functionality of a package by incorporating active substance(s) into the packaging material or the package so that it could interact with the contents and environment to extend the shelf life while maintaining the organoleptic properties of food products (Brody, Bugusu, Han, Sand, & McHugh, 2008; Realini & Marcos, 2014). Active packaging can exist in the forms of antimicrobials, antioxidants, and oxygen scavengers, as well as modified atmosphere packaging (MAP), which is the modification of the internal gaseous composition of a package.

In addition to active packaging, sustainable and biodegradable packaging have garnered significant interest among consumers due to the increase in environmental awareness. This is due to the alarming growth of global petrochemical-based plastic demand and production in the recent decades (Piñeros-Hernandez, Medina-Jaramillo, López-Córdoba, & Goyanes, 2017).

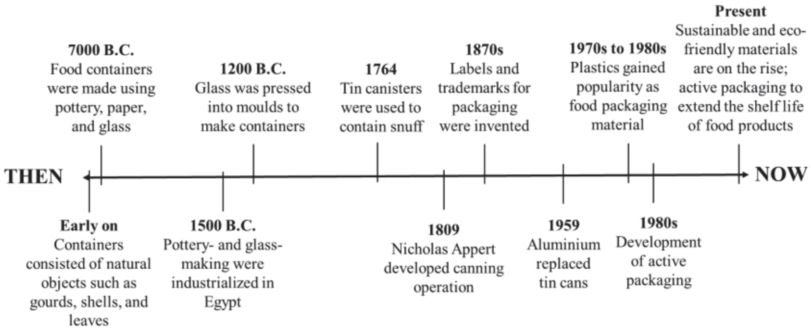

Figure 1.1 : A brief history of food packaging.
(Source: Berger & Welt, 2005; Frank et al., 2012; Robertson, 2019)

In fact, the worldwide production of petrochemical-based plastics in 2019 was 368 million tonnes, and packaging materials comprised the highest fraction of the total plastic demand, at approximately 40% (Figure 1.2) (*Plastics – the Facts 2020*, 2020). However, only 42% of the collected post-consumer packaging waste was recycled, according to a study conducted in Europe (*Plastics – the Facts 2020*, 2020). To make matters worse, a large fraction of the polymers used in the packaging industry are from non-renewable sources and therefore, exacerbating the environmental pollution. Biodegradable films produced from biopolymers such as polysaccharides, proteins, and waxes are promising alternatives to replace the plastic non-degradable films that presently plague the environment (dos Santos Caetano et al., 2018; Medina Jaramillo, Gutiérrez, Goyanes, Bernal, & Famá, 2016).

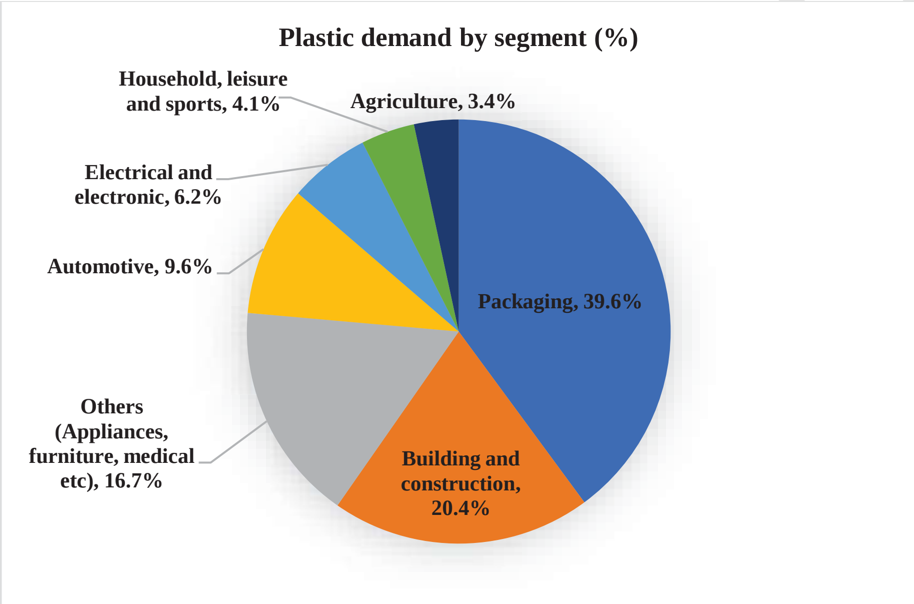

Figure 1.2 : Worldwide plastic demand by segment in 2019.
(Source: *Plastics – the Facts 2020*, 2020)

Chitosan (CS) is a hydrophilic polysaccharide derived from the partial deacetylation of chitin, which is usually found in the exoskeleton of crustaceans (Islam, Shahruzzaman, Biswas, Sakib, & Rashid, 2020). Chitin is also present in some microorganisms such as bacteria, fungi, protozoan, and algae species, making it the second most abundant polysaccharide after cellulose (Lizardi-Mendoza, Argüelles Monal, & Goycoolea Valencia, 2016). Several chitin sources have been used for the production of CS, but the most common sources of chitin are the processing waste of shellfish (Gomes, Paschoalin, & Del Aguila, 2017). As a widely distributed and environmentally friendly biopolymer, CS has attracted significant attention as a packaging material due to its nontoxicity, biocompatibility, excellent film-forming ability, as well as antimicrobial and biodegradable properties (Ahmed, Mulla, Arfat, & Thai, 2017; Leceta, Guerrero, Ibarburu, Dueñas, & Caba, 2013; Zhong & Xia, 2008). In comparison to chitin, the amino and hydroxyl groups on CS facilitates the modification of CS by chemical reactions

(Lizardi-Mendoza et al., 2016), which enables researchers to tailor its properties for its usage.

The incorporation of nanoparticles into food packaging materials is known to improve packaging performances, such as mechanical properties as well as barrier properties in terms of light, water vapour, and gas (Souza & Fernando, 2016). One of the nanomaterials that is highlighted is graphene. Graphene is carbon in the form of a single-layered graphite. It possesses the thickness of one atomic layer and the carbon atoms are arranged in a honeycomb lattice structure in the sp^2 hybridization state. This two-dimensional structure was studied immensely by the theoreticians. The band structure of graphite was first calculated by Canadian physicist P. R. Wallace in 1947 (Wallace, 1947) and it was assumed not to exist freely until more than half a century later, when plane graphene was isolated by Andre Geim and Konstantin Novoselov from University of Manchester in 2004, which won them a Nobel Prize in 2010 (*The Nobel Prize in Physics 2010,* 2010).

Due to graphene's remarkable properties such as mechanical strength, flexibility, thermal and electrical conductivity, as well as high specific surface area, there have been high expectations for graphene to be incorporated into composite materials, as well as in biological applications (Mitura & Zarzycki, 2018). In addition, graphene also exhibits antimicrobial and antioxidant activities (Baali, Khecha, Bensouici, Speranza, & Hamdouni, 2019; Lalwani, Agati, Mahmud, & Sitharaman, 2016; Rajeswari & Prabu, 2018; Tayade, Borse, & Meshram, 2019), which makes it fitting as a component in active packaging. However, the bulk synthesis of defect free graphene sheets remains a challenge and is the greatest obstacle to commercialization (Merritt, Wan, Shollock, & Patole, 2018; Potts, Dreyer, Bielawski, & Ruoff, 2011). In addition, due to its highly hydrophobic properties, the direct incorporation of graphene into biopolymers in an aqueous solution can be difficult.

To overcome these problems, reduced graphene oxide (rGO), which is structurally similar to graphene, could be incorporated into the composite. The rGO can be extracted through Hummers' method, which involves the oxidation of graphite to graphene oxide (GO), followed by a reduction process into rGO (Figure 1.3). During the oxidation of graphite, the structure of GO is endowed with reactive oxygen-containing functional groups at the basal planes and edges of the GO sheets, which facilitates the exfoliation process into single layer GO. This also allows the interactions between GO and the functional groups on hydrophilic polymers through hydrogen bonds, covalent bonds, and electrostatic interactions, thus improving the dispersion of GO within the polymeric matrix (Kumar & Koh, 2014; Yan et al., 2017).

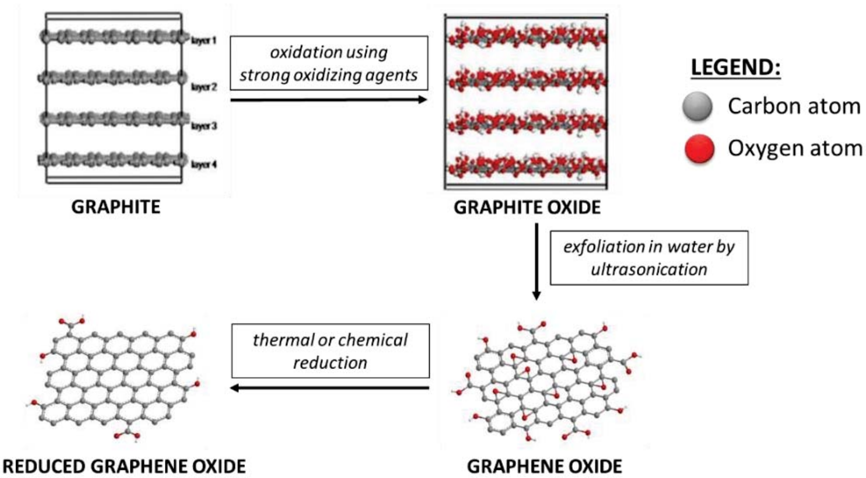

Figure 1.3 : Synthesis of graphene oxide (GO) and reduced graphene oxide (rGO) from graphite.
(Source: Naderi, Norouzi, Ganjali, & Gholipour-Ranjbar, 2016; Yuan et al., 2017)

The rGO can be incorporated into biopolymers via two approaches. The first approach involves the reduction of GO prior to incorporation, through methods such as microwave irradiation, chemical reductants, or thermal reduction. The second approach is in-situ reduction, where the reduction of GO occurs after the composite is cured. In this study, in-situ reduction was chosen due to the simplicity and safeness of this method as well as to avoid any possible aggregation of rGO in the polymeric matrix.

Besides the incorporation of nanofillers to improve the properties of biopolymeric composites, crosslinking is also an effective means to stabilize the structure of the polymeric network through the formation of a three-dimensional network of linked polymer chains. This usually results in improvements of the mechanical and barrier properties, as well as the stability of the material in water (Garavand et al., 2017; Mohamed, Mohd, Nurazzi, Siti Aisyah, & Mohd Fauzi, 2017; Nataraj, Sakkara, Meghwal, & Reddy, 2018). Depending on the properties of the crosslinking agent and the types of functional groups involved, crosslinking can be categorized into chemical (covalent), physical (ionic), and enzymatic crosslinking. Due to the ability of the amino groups in CS to be cationized in acidic media (Lizardi-Mendoza et al., 2016), physical crosslinking of CS can be effectively performed using negatively charged ions such as citrates (Li et al., 2018) as well as anionic molecules such as phosphate-bearing groups (Liang, Wang, & Chen, 2019).

In this study, the effectiveness of the composites in extending the shelf life of palm oil-based margarine was evaluated. Margarine was developed by a French chemist, Hippolyte Mège Mouriès, when Emperor Napoleon III demanded a cheaper butter alternative for the army. In the 1970s, scientists discovered that a diet high in saturated fat increased the level of low-density lipoprotein (LDL) cholesterol and decreased the level of high-density lipoprotein (HDL) cholesterol in the blood. This has caused many consumers to shift from butter to margarine, resulting in a spike in margarine's demand

(Morris & Vaisey-Genser, 2003). In terms of composition, margarine is a water-in-oil emulsion which is made mainly of refined vegetable oils from sunflower, soybean, olive, or palm oil. Commercial margarine also contains additives such as emulsifier, salt, antioxidant(s), flavouring agent(s), and vitamins. In addition to enhancing the flavour and texture of food, margarine contributes nutritionally to our diet by being a source of fat-soluble vitamins such as vitamins A and D. With the advancement of research and technologies, more brands are now offering margarines that have a low trans fatty acids content as well as reduced fat and energy (Morris & Vaisey-Genser, 2003).

Concisely, CS is a valuable and potential material to be developed as a packaging material due to its excellent film-forming properties, oxygen barrier properties, as well as its abundance in nature. This can also reduce the waste generated from the shellfish processing industries. Various nanoparticles have been incorporated to enhance the properties of a CS film as food packaging material. Graphene is a promising nanofiller because of its remarkable mechanical strength, antimicrobial and antioxidant properties, as well as a high specific surface area. However, the lack of functional groups on graphene limits its interaction with CS. Therefore, rGO can be incorporated instead of pristine graphene. This involves the addition of the hydrophilic GO into CS, followed by an in-situ thermal reduction process. Besides the addition of nanofiller, the crosslinking of a CS has also been reported to improve its mechanical and barrier properties. However, a number of these studies focused on the synthesis of CS nanoparticles or beads (Babakhani & Sartaj, 2020; Hosseini, Soleimani, & Nikkhah, 2018; Jafari, Rad, Baharfar, Asghari, & Esfahani, 2020; Pan et al., 2020; Sang et al., 2020). On top of that, many studies which reported the crosslinking of CS films focused on the application on heavy metal removal from wastewater (Luna et al., 2019), as well as drug delivery and tissue engineering (Arteche Pujana, Pérez-Álvarez, Cesteros Iturbe, & Katime, 2013; Cho et al., 2016; Gierszewska & Ostrowska-Czubenko, 2016a). The effect of ionic crosslinking using sodium tripolyphosphate (TPP) and trisodium citrate (CIT) on the mechanical, physical, and antioxidant properties of chitosan-graphene oxide (CSGO) composite and its feasibility as a food packaging material, specifically for margarine, have yet to be investigated.

The development of a CSGO composite film material with improved mechanical strength, radical scavenging properties, as well as high light, water vapour, and oxygen barriers can be a promising packaging material for margarine to slow down the rancidification and therefore, extending its shelf life.

1.2 Problem statement

As a film packaging material, the strong hydrophilicity of CS can result in a compromised mechanical strength under humid environments (Elsabee & Abdou, 2013; Han, Yan, Chen, & Li, 2011), which often leads to compromised gas and water vapour barrier. The incorporation of graphene into CS is anticipated to significantly reinforce the material. However, pristine graphene is strongly hydrophobic which makes the incorporation into hydrophilic biopolymers such as CS, extremely challenging. GO is an inexpensive source to produce rGO, which shares comparable qualities with graphene (Gupta, Sharma, Singh, Arif, & Singh, 2017).

It is understood that the oxidation degree of GO can be controlled by the concentration of the oxidants, such as $KMnO_4$, during the oxidation of graphite. Due to the abundance of oxygen-containing functional groups on its structure, a highly oxidized GO, which is synthesized using a graphite:$KMnO_4$ ratio of at least 1:6 (Marcano et al., 2010), is anticipated to incorporate homogeneously into a hydrophilic polymer such as CS, in comparison to GO with a lower oxidation degree. In addition, the epoxide groups in GO was shown to readily react with primary amine groups (Han et al., 2011). Due to the abundance of amino groups in CS, GO is expected to incorporate homogeneously into the polymeric network of CS. Nevertheless, the effect of GO oxidation degree on the incorporation into the CS polymeric matrix, as well as the effects on the structural and physical properties of the composite films as food packaging, are yet to be explored.

Previously, different sonication time have been used for the exfoliation of GO which ranged from 10 to 120 min (Liu et al., 2016; Meng, Ye, Coates, & Twigg, 2018; Pan, Wu, Bao, & Li, 2011; Wu, Wang, He, Zhang, & Zhang et al., 2017). A longer sonication time is expected to produce GO with a higher surface area-to-volume ratio, which will be more effective as a nanofiller (Cai et al., 2017; Tang, Ehlert, Lin, & Sodano, 2012). However, sonication can also damage and fragment the GO sheets (Dreyer, Park, Bielawski, & Ruoff, 2010; Parades, Villar-Rodil, Martínez-Alonso, & Tascón, 2008). In addition, the extremely high temperature resulting from the implosion of cavitation bubbles might thermally reduce GO, which could hinder the homogeneous incorporation into a hydrophilic polymer, such as CS.

The high hydrophilicity of GO is predicted to increase the water affinity of the composite (Yoo, Shin, Yoon, & Park, 2014). Therefore, the CSGO composite has to be treated in order to reduce its hydrophilicity and to improve its mechanical strength as well as oxygen and water vapour barrier properties under humid conditions. This could be achieved by the reduction of GO or the crosslinking process of the composite films to reduce the available functional groups.

Different temperatures have been applied for in-situ reduction of GO in previous studies, ranging from to 50 to 200 ºC (Grande et al., 2017; Meng et al., 2016; Olowojoba et al., 2016; Toselli et al., 2015). However, different temperatures will cause a different saturation of oxygen-containing functional groups on the resulting GO nanosheets, which eventually influences the structural, mechanical, optical, and water resistant properties of the composites, as reported by Meng et al. (2016). Therefore, the optimum temperature for the in-situ thermal reduction of GO and its effects on the CSGO composites shall be determined.

Crosslinking has been used to effectively modify and improve the properties of CS. However, gelation occurs almost immediately once the crosslinking agents are introduced into the film-forming solution (Lin, Gu, & Cui, 2019; Nataraj et al., 2018; Yan et al., 2015), which makes it impossible to pour the solution onto the film-casting surface. However, this can be avoided by introducing the crosslinking agents after the films were cast, dried, and peeled.

The oxidative rancidity of margarine is often accelerated by heat, ultraviolet light, and oxygen (Riaz & Rokey, 2012). Therefore, the packaging material of the margarine can be improved in terms of its light and oxygen barrier. At present, many of the commercially available margarine in Malaysia adapt a polypropylene (PP) tub packaging (Figure 1.4). In fact, the demand for PP was one of the highest in the packaging sector along other resins such as low-density polyethylene (LDPE), high-density polyethylene (HDPE), and polyethylene terephthalate (PET) (*Plastics – the Facts 2020*, 2020). Unfortunately, PP is one of the least recycled post-consumer plastics which has a recycle ratio of only ~0.6% (Pavlík, Pavlíková, & Záleská, 2019). The huge volume of post-consumer waste has generated large amounts of urban solid residues (Vieira Ramos, Reis, Grafova, Grafov, & Monteiro, 2020). Therefore, a biodegradable active packaging with antioxidant properties as well as low light transmittance and oxygen permeability, can be a promising alternative to current petrochemical-based packaging for margarine.

Figure 1.4 : Examples of margarine brands sold in Malaysia that are packaged in polypropylene tubs.

1.3 Objectives of the study

The aim of this study is to develop an antioxidant active packaging for margarine using chitosan (CS) with improved structural and physical properties, as well as water, light, and oxygen barrier through the incorporation of graphene oxide (GO) as well as heat and crosslinking treatments. There are four specific objectives in this study;

1. To prepare and evaluate the effect of oxidation degrees of GO on the structure and physical properties of CS films.
2. To evaluate the effect of sonication time and heating temperature on the structural and physical properties of chitosan-graphene oxide (CSGO) nanocomposite films.
3. To investigate the effect of trisodium citrate (CIT) and sodium tripolyphosphate (TPP) crosslinkers on the properties of CSGO nanocomposite films.
4. To evaluate the effect of CS and GO concentrations on the physicochemical properties of active packaging and their effects on storage stability of palm oil-based margarine.

The general research flow of this study, from Objective 1 to Objective 4, is summarized in Figure 1.5.

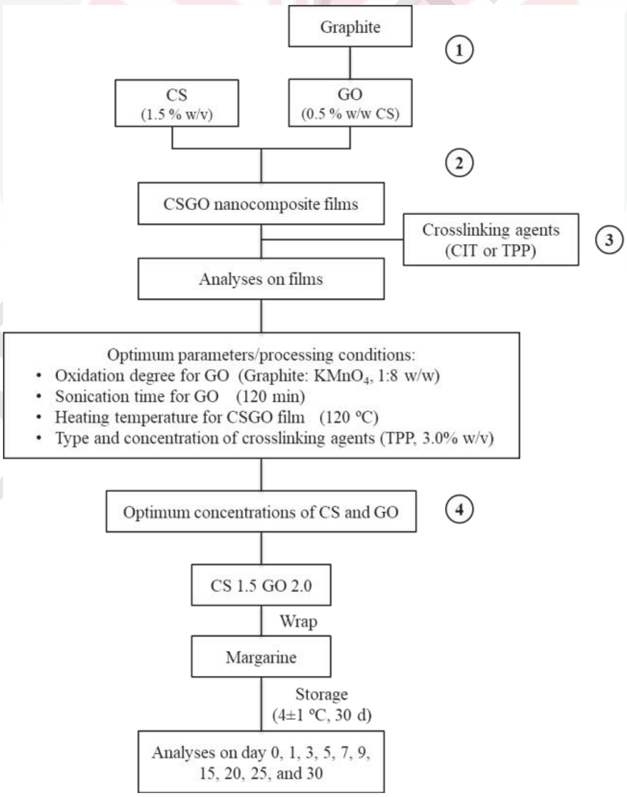

Figure 1.5 : Research flow of the study.

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