



**CHARACTERIZATION OF BROILER CHICKEN HEAD GELATIN FOR  
MICROCAPSULES USING CROSSLINKED GELATIN-SAGO STARCH  
COMPLEX COACERVATES**

By

**EE SHU CHEE**

**Thesis Submitted to School of Graduate Studies, Universiti Putra Malaysia, in  
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Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfilment of the requirement for the degree of Doctor of Philosophy

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**December 2022**

**Chair : Professor Faridah Abas, PhD**  
**Faculty : Food Science and Technology**

Malaysia is a surplus poultry producing country, and an immense quantity of poultry by-products are generated, most of which are discarded or underutilized. Chicken heads could be a potential source of gelatin as it is rich in protein. Gelatin extraction methods with combination of alkaline-acid pretreatment (E1), acid pretreatment (E2) and alkaline pretreatment (E3) were studied to extract the gelatin from broiler chicken heads. Methods E1 and E2 produced high bloom Type A gelatin (> 300 g) with < 1% ash content, high gelling (25.8-26.0 °C) and melting temperatures (30.8-32.3 °C), good functionality and physical appearance. Compared to bovine gelatin, gelatins of E1 and E2 had higher viscous ( $G''$ ) and elastic modulus ( $G'$ ) values on cooling and heating. FTIR spectra indicated different degree of structural denaturation in gelatins. Both methods E1 and E2 yielded higher quality gelatin than E3. Yet, the combination of alkaline-acid pretreatment in E1 was the most effective extraction method to obtain high quality gelatin from chicken heads. Next, the physicochemical properties of gelatin extracted from the chicken head at 60, 75 and 90 °C for 3 and 6 h, respectively were determined. An increase in extraction temperature (ETE) and extraction time (ETI) contributed to higher yield and bloom. At 90 °C, gelatin bloom and viscoelastic properties began to drop. ETE was highly correlated with yield ( $r = 0.954$ ). Gelatin extracted at 75 °C exhibited superior properties with bloom strength of > 309 g, high in  $G'$ ,  $G''$ , gelling (27-28 °C) and melting (33-34 °C) temperatures with great viscoelastic properties. All gelatins were of type A with  $\alpha$ -chains as major protein. FTIR bands indicated different degrees of structural change. Glycine was the main amino acid in G75/6 (20.69%) with total imino acid of 23.19%. Regression models were significant ( $p < 0.05$ ) and highly fitted for yield and  $a^*$  ( $R^2 > 0.9779$ ). Present findings suggest the feasibility to extract high quality gelatin from the chicken head by manipulating ETE and ETI. Complexes formation from chicken head gelatin (CHG) and sago starch (SA) was investigated as a function of pH (8.0-2.0), CHG-SA ratio (1:5-5:1, w/w), biopolymer concentration (0.1-0.9%, w/v) and sago starch concentration (0.1-0.9%, w/v). The optimum conditions for CHG-SA coacervation (1:3, w/w) were identified to be pH 6.02, achieving maximum turbidity and zero  $\zeta$  potential value. The increased in total biopolymer (0.1%-0.9%) and SA

concentration increase the turbidity value. The p<sub>Hopt</sub> increased as the CHG-SA mixing ratio (1:5-1:1) increased. The CHG-SA complexes had higher amide I peak intensity than CHG in FTIR analysis, indicating the formation of coacervates. Following this, CHG-SA complex coacervates were crosslinked with tannic acid (TA) and employed as the shell material to produce Citronella oil. The secondary structure of gelatin was observed to change in crosslinked CHG-SA coacervates, as evidenced by a shift in the amine spectrum due to hydrogen bonding with TA. Crosslinking improved the thermal stability and particle size of the coacervates due to the hardening effect. Crosslinked coacervates displayed higher peak intensity in X-ray diffraction analysis suggesting enhanced intermolecular bonding (increase in gelatin helix) after crosslinking. Citronella oil microcapsules prepared from crosslinked CHG-SA coacervates had an average particle size of 78 μm with an encapsulation efficiency of 68% and were thermally stable up to 315 °C. Overall, high quality gelatin could be extracted from chicken heads, and this gelatin could form complex with sago starch and to create CHG-SA coacervates. Such coacervates can be used as a wall material to encapsulate citronella oil, preserving its bioactivity for further food applications.

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

**PENCIRIAN GELATIN KEPALA AYAM BROILER UNTUK PENYEDIAAN  
MIKROKAPSUL MENGGUNAKAN KOMPLEKS KOASERVASI GELATIN-  
KANJI SAGU DISAMBUNG SILANG**

Oleh

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**Disember 2022**

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Industri ternakan ayam berskala besar di Malaysia telah menghasilkan banyak produk sampingan ayam yang kebanyakannya dibuang atau kurang digunakan. Kepala ayam yang kaya dengan protein amat berpotensi untuk dijadikan sumber gelatin. Kaedah pengekstrakan gelatin yang melibatkan penggunaan alkali-asid (E1), asid (E2) dan alkali (E3) telah dikaji untuk mengekstrak gelatin daripada kepala ayam. Kaedah E1 dan E2 menghasilkan gelatin Jenis A dengan kekuatan bloom yang tinggi ( $> 300$  g), kandungan abu  $< 1\%$ , suhu gel ( $25.8\text{-}26.0$  °C) dan suhu lebur ( $30.8\text{-}32.3$  °C) yang tinggi, mempunyai fungsi gelatin dan penampilan fizikal yang baik. Gelatin E1 dan E2 mempunyai nilai modulus likat ( $G''$ ) dan modulus elastik ( $G'$ ) yang lebih tinggi daripada gelatin lembu semasa process penyejukan dan pemanasan. Spektra FTIR menunjukkan struktur gelatin mengalami tahap denaturasi yang berbeza. Kaedah E1 dan E2 menghasilkan gelatin yang lebih berkualiti daripada E3. Walau bagaimanapun, kaedah kombinasi alkali-asid (E1) dikenalpasti sebagai kaedah pengekstrakan terbaik untuk menghasilkan gelatin yang berkualiti tinggi daripada kepala ayam. Seterusnya, pengekstrakan gelatin daripada kepala ayam telah dijalankan pada  $60$ ,  $75$  dan  $90$  °C selama  $3$  dan  $6$  jam dan sifat fizikokimia gelatin tersebut ditentukan. Peningkatan masa (ETI) dan suhu ekstraksi (ETE) meningkatkan hasil ekstraksi dan kekuatan bloom gelatin. Akan tetapi, kekuatan bloom dan sifat viskoelastik mula menurun pada  $90$  °C. ETE berkorelasi kuat dengan hasil ekstraksi ( $r = 0.954$ ). Gelatin yang diekstrak pada suhu  $75$  °C mempamerkan sifat superior dengan kekuatan bloom  $> 309$  g, mempunyai nilai  $G'$ ,  $G''$ , suhu gel ( $27\text{-}28$  °C) dan suhu lebur ( $33\text{-}34$  °C) yang tinggi serta menunjukkan sifat viskoelastik yang baik. Semua gelatin ekstraksi adalah Jenis A dan mempunyai rantai  $\alpha$  sebagai protein utama. FTIR menunjukkan struktur gelatin mengalami tahap perubahan yang berbeza. Glycine merupakan asid amino utama dalam gelatin G75/6 (20.69%) dengan kandungan asid imino sebanyak 23.19%. Model regresi adalah signifikan ( $p < 0.05$ ) dan fit untuk hasil ekstraksi dan nilai  $a^*$  ( $R^2 > 0.9779$ ). Kajian ini mencadangkan kebolehlaksanaan untuk mengekstrak gelatin berkualiti tinggi daripada kepala ayam dengan memanipulasikan ETE dan ETI. Dalam kajian ketiga, pembentukan kompleks daripada gelatin kepala ayam (CHG) dan kanji sago (SA) telah dikaji atas faktor pH ( $8.0\text{-}2.0$ ), nisbah CHG-SA

(1:5-5:1, w/w), kepekatan biopolimer (0.1-0.9%, w/v) dan kepekatan kanji sago (0.1-0.9%, w/v). Keadaan optimum untuk koaservasi CHG-SA (1:3, w/w) dikenal pasti pada pH 6.02, mencapai turbiditi maksimum dan nilai potensi  $\zeta$  sifar. Nilai turbiditi meningkat apabila jumlah kepekatan biopolimer (0.1-0.9%) dan kepekatan SA meningkat. Nilai pH<sub>opt</sub> meningkat seiring dengan peningkatan nisbah pencampuran CHG-SA (1:5-1:1). Kompleks CHG-SA mempamerkan intensiti puncak amida I yang lebih tinggi berbanding CHG dalam analisis FTIR, ini menunjukkan pembentukan koaservasi. Seterusnya, kompleks koaservasi CHG-SA disambung silang dengan asid tannik (TA), dan digunakan sebagai dinding untuk memikroenkapsulasikan minyak Citronella. Penyilangan mengubah struktur sekunder gelatin dalam koaservasi CHG-SA, seperti yang dibuktikan dalam peralihan amina dalam spektra FTIR akibat ikatan hidrogen dengan TA. Penyilangan meningkatkan kestabilan terma dan ukuran partikel akibat kesan pengerasan proses penyilangan. Analisis difraksi sinar-X menunjukkan koaservasi CHG-SA yang ditaut silang mempunyai intensiti puncak yang lebih tinggi akibat peningkatan helik gelatin dan ikatan antara molekul selepas proses penyilangan. Mikro kapsul minyak Citronella yang disediakan daripada koaservasi CHG-SA disambung silang mempunyai ukuran partikel purata 78  $\mu\text{m}$  dengan kecekapan pengkapsulan 68% dan stabil sehingga 315 °C. Secara keseluruhannya, gelatin berkualiti tinggi boleh diekstrak daripada kepala ayam, dan gelatin ini boleh dikomplekskan dengan kanji sago untuk membentuk koacervasi CHG-SA. Koacervasi ini boleh digunakan sebagai bahan dinding untuk mengkapsulasi minyak Citronella, mengekalkan bioaktivitinya untuk aplikasi makanan selanjutnya.

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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 ABBREVIATIONS

ANOVA	Analysis of Variance
ATR	Attenuated Total Reflectance
CAGR	Compound Annual Growth Rate
CHG	Chicken head gelatin
CHG-SA	Chicken head gelatin-sago starch
ETE	Extraction temperature
ETI	Extraction time
FTIR	Fourier Transform Infrared Spectroscopy
GMIA	Gelatin Manufacturers Institute of America
HCl	Hydrochloric acid
HPLC	High Performance Liquid Chromatography
Hyp	Hydroxyproline
pI	Isoelectric point
NaCl	Sodium chloride
NaOH	Sodium hydroxide
OECD	The Organization for Economic Cooperation and Development
Pro	Proline
SA	Sago starch
SDS-PAGE	Sodium dodecyl-sulfate polyacrylamide gel electrophoresis
SEM	Scanning Electron Microscope
TA	Tannic acid
TGA	Thermal Gravimetric Analysis
XRD	X-Ray Diffraction

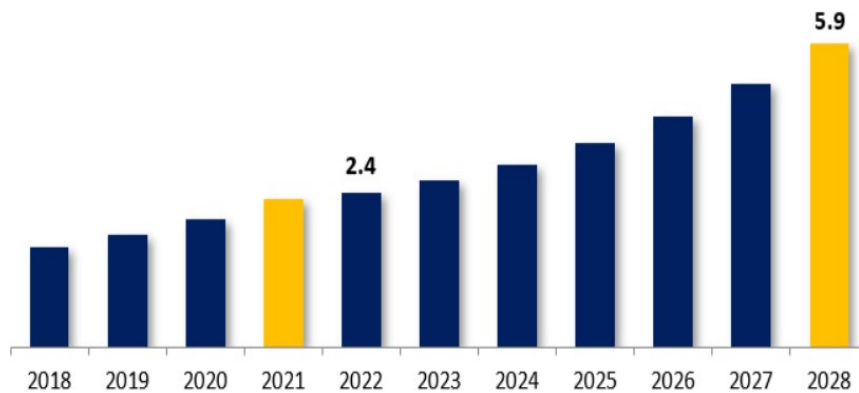
# CHAPTER 1

## INTRODUCTION

### 1.1 Background and problem statement

What makes gelatin a popular ingredient in the food industry? It is its versatility, unique thermo-reversible properties and has multiple functions compared to other biopolymers. Gelatin (Latin: gelatos meaning “stiff”) is polypeptides obtained by the partial hydrolysis of collagen from animal skin, bones, and white connective tissue. Gelatin is practically tasteless and odourless brittle solid in a faintly yellow color and it has ability to form thermos-reversible gels. It is mainly used in food such as confections, reduced calorie products, baked goods, dairy products, meat and sausages products for its gelling, foaming, emulsifying properties, and other unique functionalities such as texturizer, thickener, water binder, mouthfeel provider, fat reduction, refining agent, colloid stabilizer, and encapsulating properties. In the pharmaceutical, cosmetic and medical industries, gelatin is used to make hard and soft capsules, protein-enriched body-building food products, carbohydrate reducers, dietary supplements, edible films and coatings, hair care products, matrix for implants, plasma expanders, wound dressing, drug delivery microspheres and so on (Karim & Bhat, 2009; Abedinia et al., 2020).

The Global Gelatin Market was valued at USD 2.4 billion in 2022 and is expected to grow to USD 5.9 billion by 2028, at a CAGR of 4.3 % (Figure 1.1). Conventional commercial gelatin sources are mostly manufactured from bovine and porcine bones and hides. Due to the long life cycle of the two conventional sources and increasing global demand in the food and pharmaceutical industries, there has been an aggressive search for alternative gelatin sources with equal or superior properties. Chicken heads could fill in this additional demand of gelatin. This portion of poultry are generally regarded as useless, had been discarded by-poultry processing plants and their uses are more to niche market such as being rendered into poultry by-products meal for feeding farmed fish and shrimp in Malaysia. This underutilized proteinaceous poultry by products have great potential for value-added food applications.



**Figure 1.1: The global gelatin market by value (USD Billion), 2018-2028** (Source: BlueWeave Consulting, 2022)

Chicken heads make up around 2% of the body (skin, comb, wattle, cartilages, and bones), although not consumed by human, is an appropriate and promising source for gelatin production due to their high collagen content. Production of gelatin from chicken heads could provide a sustainable gelatin supply as poultry has much shorter production cycle that allows production benefits compared to bovine and porcine. It is economically viable to extract gelatin from chicken head given the projected rapid growth in the industries in years to come with the continuous supply of raw material from poultry processing. Poultry processing industry is possibly one of the world's fastest growing agro-food sectors, with the world poultry meat production is anticipated to reach 141 million metric tonnes by 2028 (OECD/FAO, 2021). At the same time, approximately 22-30% or 40 million metric tonnes of poultry by products (including head, viscera, blood and feathers) were generated. Furthermore, gelatin from poultry has almost no issue with the dietary and religious concern as long as those poultry are raised with proper farming practice and slaughter according to the halal and good slaughtering procedure.

Proteins and polysaccharides are two of the most used essential ingredients that contribute greatly to the functional properties of a food system. Complexation between proteins and polysaccharides through electrostatic interactions lead to formation of complex coacervate or liquid-liquid phase separation, and are influenced by processing conditions such as pH, ionic strength, protein to polysaccharide ratio and crosslinking agent (Devi et al., 2017). Gelatin-polysaccharides complex coacervation system is very attractive due to their availability, biocompatibility, biodegradability and safe characteristics. The preparation of protein-polysaccharide complexes with enhanced functionalities such as improved thermal stability, emulsifying and rheological properties provide opportunities for the development of new ingredients for the food industry. Complexes can be used as food stabilizer, emulsifier, fat replacer, cosmetic and gaining research interest for microencapsulation. Gelatin-polysaccharide complexes was employed as wall material for encapsulating hydrophobic flavor oils into microspheres through coacervation process (Lengyel et al., 2019). During this process, crosslinking reagent such as tannic acid was added to harden and strengthen the structure of

microcapsule walls instead of conventional aldehydes reagents that is toxic and undesirable for food use.

The general objective of this study was to produce gelatin from the chicken head with different extraction methods and conditions. Proper study of the extraction procedures led to the extraction of good quality gelatin from chicken head. The extracted gelatins are stabilized and characterized. Gelatin extraction methods were optimized for quality and yield improvement. Findings on gelatin properties and gelatin recovery prediction may benefit the further industrial sectors. In the second stage, the complexation of extracted gelatin with sago starch was performed under the influence of pH, ratio and concentration of biopolymers. The effect of modification is investigated and the optimum condition for complex coacervation of chicken head gelatin-sago starch (CHG-SA) were identified. Gelatin behaves cationically in coacervation due to its polypeptide structure that favors interactions with wide range of water-soluble anionic polymers to form complex. Sago starch is an underutilized crop with promising industry potential due to its high yield and inexpensive, which makes it worthwhile for exploitation. The fabricated CHG-SA complex was then crosslinked with tannic acid (TA) and employed as wall material to produce citronella oil microcapsules. The resulting CHG-SA complex coacervates and citronella oil microcapsules were characterized in terms of their protein conformation, microstructure and thermal properties.

## 1.2 Objectives

The objectives of this study were as below,

1. To extract and determine the characteristic of gelatin from chicken heads by acid and alkaline extraction methods.
2. To determine the combined effect of temperature-time using warm water rendering on the properties of extracted chicken head gelatin.
3. To investigate the formation of complex coacervation from gelatin and sago starch as a function of pH, biopolymer ratio and concentration.
4. To investigate the effect of tannic acid crosslinking on gelatin and sago starch complex coacervates
5. To prepare citronella oil microcapsule from tannic acid-crosslinked gelatin and sago starch complex coacervates

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