# ANTIMICROBIAL ACTIVE AND SMART FILM WITH SYNERGICAL EFFECT OF THYMOL AND COLOUR INDICATOR

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# ANTIMICROBIAL ACTIVE AND SMART FILM WITH SYNERGICAL EFFECT OF THYMOL AND COLOUR INDICATOR

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requirements for the award of the degree of
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Specially dedicated to my lovely and supporting parents, Khairuddin Mahmud and
Zainab Likam and family members, Khairuza Wawiyah and family,
Khairuza Izyani and family, Rozieana, Nur Bahariyah and family and
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## ABSTRACT

Active and smart packaging is a promising form of food packaging that offers a great economical potential. This is due to consumer demand for a packaging that accommodate a hectic way of life. An antimicrobial packaging with pH colour indicator (AMI) can be made by incorporating suitable antimicrobial (AM) agent and colour indicator into food package matrices whilst applying a bio switch concept to inhibit the pathogenic microorganisms and respond automatically to changes (external stimuli) in the environment. The present work aimed to develop the formulation of hydroxyethylcellulose (HEC)/wheat-starch based AMI film in which the active compound, thymol (0.5, 1, 1.5, 2, and 2.5% w/w) and 50:50% w/w bromothymol blue and methyl red (as the colour indicator) were incorporated into the polymeric material. A solution casting method was used in the film preparation while thymol and colourant were incorporated prior to casting. In order to determine the film characteristics, physical (scanning electron microscopy (SEM) and tensile test), chemical (Fourier transform infrared (FTIR)), and thermal properties (thermogravimetric analysis, differential scanning calorimetry) tests were carried out. Effects of adding thymol into the film base were studied in order to improve the antimicrobial spectrum activity based on measured agar diffusion assay. The distributions of inhibitory effect of the film were determined towards different types of microbial contamination in terms of Gram-positive bacteria, Gram-negative bacteria and fungus. The concentration of thymol as antimicrobial agent added into the film was optimized and modeled in relation to pH changes due to food microbial spoilage activities. The effect of thymol showed a range of microbial inhibition zones of 16.3 - 26.4% and 22.1 - 39.9% towards *E. coli* and *B. subtilis*, respectively. Whilst, a lower inhibition zone of 0.4 - 5.1% was demonstrated for fungus *A. niger*. The mechanical properties of the films were improved by 60.3% with an optimum tensile strength at thymol concentration of 1.5% w/w. Chemical interactions were studied by means of FTIR spectroscopy and SEM was used to verify the surface and cross section of the film matrix. The results demonstrated that thymol could be successfully added into the film without changing the main packaging properties. Direct incorporation of methyl red and bromothymol blue into film was suitable technique for making AMI packaging film. The combination of the indicators showed a distinct colour changes between pH 6 to pH 8 where most food spoilage occur. It can be concluded that the film properties are retained chemically whereas mechanical properties, strength, flexibility and function of the HEC/starch based AMI film are being enhanced remarkably by the incorporation of thymol and pH colour indicator.

# ABSTRAK

Pembungkusan aktif dan pintar adalah satu bentuk pembungkusan makanan yang menawarkan potensi ekonomi yang besar. Ia adalah berikutan permintaan pengguna untuk pembungkusan yang boleh membantu dalam kesibukan kehidupan. Suatu pembungkusan yang mengandungi antimikrob dengan penunjuk pH berwarna (AMI) boleh dihasilkan dengan menggabungkan agen antimikrob yang sesuai (AM) dan penunjuk warna ke dalam matriks pembungkusan dengan menggunakan konsep bio-suis bagi menghalang mikroorganisma patogen dan bertindakbalas secara automatik terhadap perubahan (rangsangan luar) dalam persekitaran. Kajian ini bertujuan membangunkan formulasi filem AMI berasaskan hidroksietilselulosa (HEC)/kanji gandum dengan sebatian aktif, timol (0.5, 1, 1,5, 2, dan 2.5% w/w) serta 50:50% w/w bromotimol biru dan metil merah (sebagai penunjuk warna) digabungkan ke dalam bahan polimer. Kaedah penuangan larutan digunakan dalam penyediaan filem di mana timol dan penunjuk warna telah digabungkan terlebih dahulu sebelum penuangan. Bagi menentukan sifat-sifat filem, ujian-ujian fizikal (mikroskop elektron pengimbas (SEM) dan ujian tegangan), kimia (inframerah transformasi Fourier (FTIR)), dan sifat haba (analisis termogravimetri, kalorimetri pengimbasan pembezaan) telah dijalankan. Kesan penambahan timol ke dalam bahan asas filem dikaji dalam usaha meningkatkan spektrum aktiviti antimikrobial berdasarkan ukuran assai agihan resapan agar. Kesan perencatan ke atas filem ditentukan terhadap jenis pencemaran mikrob berbeza iaitu bakteria Gram-positif, bakteria Gram-negatif dan kulat. Kepekatan timol sebagai agen antimikrob ke dalam filem ini telah dioptimum dan dimodelkan terhadap perubahan pH yang disebabkan oleh aktiviti kerosakan mikrobial makanan. Keberkesanan timol menunjukkan julat peratus zon perencatan sebanyak 16.3 - 26.4% dan 22.1 - 39.9% masing-masing bagi *E. coli* dan *B. subtilis*. Manakala zon perencatan pada julat yang lebih rendah iaitu 0.4 - 5.1% ditunjukkan terhadap *A. niger*. Sifat-sifat mekanikal filem didapati telah bertambah baik sebanyak 60.3% dengan kekuatan tegangan yang optimum pada kepekatan timol sebanyak 1.5% w/w. Interaksi kimia telah dikaji dengan kaedah spektroskopi FTIR dan SEM telah digunakan bagi mengesahkan sifat permukaan dan keratan rentas matriks filem. Keputusan menunjukkan timol berjaya digabungkan ke dalam filem tanpa mengubah ciri-ciri utama pembungkusan. Penambahan langsung bromotimol biru dan metil merah ke dalam filem didapati sesuai bagi menghasilkan filem pembungkus AMI. Gabungan penunjuk warna telah menunjukkan perubahan filem pembungkusan warna yang ketara antara pH 6 hingga pH 8 di mana kebanyakan kerosakan makanan berlaku. Kesimpulannya, ciri-ciri kimia filem dapat dikekalkan manakala sifat-sifat mekanikal, kekuatan, fleksibiliti dan kefungsian filem AMI berasaskan HEC/kanji berjaya disesuaikan dengan ketara melalui penambahan timol dan penunjuk pH berwarna.

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# LIST OF ABBREVIATIONS

| | | |
|---|---|---|
| AM | - | Antimicrobial |
| AMI | - | Antimicrobial with Indicator |
| ASTM | - | American Society for Testing and Materials |
| BB | - | Bromothymol blue |
| CAGR | - | Compound Annual Growth Rates |
| $CO_2$ | - | Carbon dioxide |
| DOF | - | Degree of Freedom |
| DSC | - | Differential Scanning Calorimetry |
| DTG | - | Derivatives Thermo Gravimetric |
| EDTA | - | Ethylenediaminetetraacetic acid |
| EFSA | - | European Food Safety Agency |
| EPA | - | Environmental Protection Agency |
| EVA | - | Ethylene vinyl acetate |
| FTIR | - | Fourier-transform infra-red |
| HDPE | - | High density polyethylene |
| HEC | - | Hydroyethylcellulose |
| HPLC | - | High Performance Liquid Chromatography |
| LDPE | - | Low density polyethylene |
| MS | - | Means Square |
| PE | - | Polyethylene |
| PG | - | Propylene Glycol |
| PEG | - | Polyethylene Glycol |
| PET | - | Polyethylene terephtalate |
| PP | - | Polypropylene |
| PVC | - | Polyvinylchloride |

| | | |
|---|---|---|
| PVOH | - | Polyvinyl alcohol |
| SEM | - | Scanning electron microscopy |
| $SO_2$ | - | Sulfur dioxide |
| SS | - | Sum of Squares |
| TGA | - | Thermo Gravimetric Analysis |
| TPS | - | Thermoplastic Starch |
| TS | - | Tensile strength |
| US FDA | - | United States Food and Drug Administration |
| UV | - | Ultra violet |
| YM | - | Young's modulus |

# LIST OF SYMBOLS

| | | |
|---|---|---|
| % | - | percentage |
| 3-D | - | three dimensional |
| °C | - | degree celsius |
| µm | - | micrometer |
| *a* | - | colour opponent dimension |
| *b* | - | colour opponent dimension |
| $cm^{-1}$ | - | reciprocal centimeters |
| $cm^3$ | - | cubic centimeter |
| *et al.*, | - | and others |
| etc. | - | and other things |
| G | - | gram |
| $g/cm^3$ | - | gram per cubic centimeter |
| h | - | hour |
| kV | - | kilovolt |
| *L* | - | lightness |
| mg | - | milligram |
| min | - | minutes |
| $min^{-1}$ | - | revolutions per minute |
| mL | - | mililiter |
| mm | - | milimeter |
| mm/min | - | millimeter per minute |
| Nm | - | nanometer |
| pH | - | potential of Hydrogen |
| $R^2$ | - | regression value |
| s | - | second |

| | | |
|---|---|---|
| $T_i$ | - | initial temperature |
| $T_m$ | - | melting temperature |
| $T_{max}$ | - | maximum temperature |
| w/v | - | weight per volume |
| $\Delta E$ | - | delta E |
| %T | - | Transmittance |

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# CHAPTER 1

# INTRODUCTION

## 1.1 Background

Packaging plays an important role in the whole food chain 'from the field to the consumer's table'. Furthermore, food packaging has developed strongly during recent years, mainly due to the increased demands on product safety, shelf-life extension, cost-efficiency, environmental issues and consumer convenience. Owing to increasing urban lifestyle and global population trends, the demand for packaged, frozen, and ready-to-eat food has witnessed a significant surge in recent times. With supply of exotic fruits and vegetables, meat products and frozen foods transcending geographical boundaries, the packaging industry has been focusing on development of solutions that provide maximum food security while maintaining nutritional value at competitive prices.

Packaging poses new challenges to the evaluation of its safety as compared to the traditional packaging due to its deliberate interaction with the food and/or its environment active and packaging i.e. migration of substances from packaging to food, incorrect use of the packaging due to the insufficient labeling, non-efficacious operation of the packaging, etc. (Dainelli *et al.*, 2008; Restuccia *et al.*, 2010).

Global advanced packaging technology market is witnessing a high growth due to the increase in demand for convenience food such as ready-to-cook meal, stricter food safety regulations, and demand for sustainable packaging. Growing health awareness among consumers, food wastage, and manufacturers' concern for longer shelf life of the food products and supply chain inefficiencies are the other factors fuelling the growth of the market. Further, the consumers' confidence with regards to packaged food, organized food safety regulations, threat from bioterrorism, and increased applications in ready-to-eat meals and frozen foods have created a huge opportunity for advanced packaging technology market. In 2010, the active and intelligent packaging technology held the highest growth rate, estimated at a Compound Annual Growth Rate (CAGR) of 10.5% and 12.1% from 2010 to 2015 respectively. Modified atmosphere technology accounted for largest share (approximately 54%) of the total market in advanced packaging technology (Figure 1.1). The global advanced packaging technology is estimated to grow at a CAGR of 8.2% from 2010 to 2015.

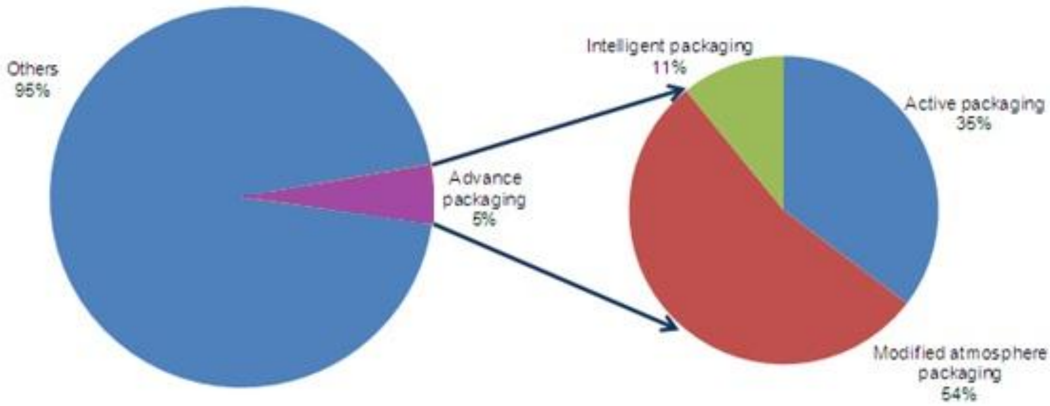


**Figure 1.1** The global advanced packaging technology is estimated to grow at a CAGR of 8.2% from 2010 to 2015.

Existing examples of smart packaging may consist of temperature indicators/sensors, applied to signal a maximum- or minimum-temperature event; time temperature integrators (TTIs), one of the most commercially applied smart packaging devices; moisture (i.e., relative humidity) sensors; and gas sensors indicating excess or shortage of requisite reactant. Biosensors evaluate biological activities such as microbial growth in spoilage indicators. Freshness indicators are declared to respond to ripeness signals. Package integrity indicators, try to ensure against contamination and radio frequency identification (RFID) are perhaps the most promising of all smart packaging concepts, but still with restrictions.

## 1.2 Problem Statement and Hypothesis

The food and beverage packaging has dramatically shifted from traditional to advanced packaging. Traditional packaging only addresses issues related to protection from external factors. However, advanced packaging interacts internally (active packaging) and externally (intelligent packaging) with the environment and enhances the visual appeal of the products. Therefore, manufacturers of food and beverages and packaging manufacturers are looking into the way the physical, chemical and microbial textures change inside the packaged food. The requirement over smart or intelligent packaging is changing rapidly due to the world awareness on environment. The concerns are not only on the materials and productions, but also on legal side related to the environmental concerns.

The general concept of bio-switch describes a system with capability to detect and respond automatically to changes (external stimuli) in the environment. For instance, the external stimulus may be a change in pH, or the presence of certain metabolites from biological activity. The bio-switch converts this stimulus into a particular functionality. Materials with ability to entrap compounds with a specific function which are released on an external stimulus from the environment are created. In order to be used in active packaging, biopolymer-based particles containing antimicrobial will only be released in the case of initial microbial contamination. The bio-switch particles monitor the releasing system by the stimulus of a microbial contamination that actively add or emit compounds *i.e.* antimicrobials, antioxidants, and preservatives to the packaged food or onto the surface of the package (Muhamad *et al*., 2008). The current work is a novel combination of active packaging technology and smart packaging concept where the stimulus of a microbial contamination is further incorporated with an indicator to signal the conditions in the packaged food.

Therefore, incorporation of thymol and colour indicators into starch based film will be expected to create synergy effect of thymol and colour indicators towards prolonging the shelf life of packed food while simultaneously capable to communicate with consumer on unexpected changes of the packed food leading to spoilage and pH changes.

## 1.3 Objectives of Study

AM starch-based packaging unites with colour indicator is a competent form of intelligent food packaging with lot of benefit; economically, environmentally, and generally give a lot of health potentials. Objectives of this research are:

1. To determine the optimum formulation of antimicrobial starch-based film HEC/Starch/Thymol (AM film),
2. To investigate the effect of antibacterial agents towards the inhibition of microbial activity,
3. To determine the optimum formulation of antimicrobial starch-based film with indicator, HEC/Starch/Thymol/Colour indicators, (AMI film),
4. To determine the effectiveness of the AMI film towards pH changes.

## 1.4 Scopes of Study

In order to achieve the objectives of this study, the performed work included the following scopes. In this study, starch was used as packaging material incorporated with thymol as antimicrobial agents in order to develop the formulation of HEC/Starch/Thymol (AM) film. Whilst, the formulation was further improved with the additional of methyl red and bromothymol blue as the colour indicators in order to develop the HEC/Starch/Thymol/Indicators (AMI) film. Solutions of starch and HEC with different concentration of thymol ranges from 0.5 – 2.5 % (w/v) were blended to formulate the synergy AM film using the combination of HEC/Starch/Thymol. Whereas, the film solutions of HEC/Starch/Thymol were improved with the incorporation of a constant composition of colour indicators to formulate the synergic AMI films with the combination of HEC/Starch/Thymol/colour indicators.

Antimicrobial films formulated were then characterized based on mechanical, physical, thermal and chemical properties. Besides that, an evaluation of the effectiveness of the prepared AM and AMI films were evaluated based on the antimicrobial activity against Gram-positive (*B. subtilis*) and Gram-negative (*E. coli*) bacteria and fungus (*A. niger*). Lastly, the scope of study included the determination on the factors influencing the colour changes of indicator due to changes of pH.

## 1.6 Outline of the Thesis

This thesis consists of six chapters. Chapter 1 is the introduction of the research, research problem and hypothesis, the objectives and the scope of the study. Chapter 2 presents the literature review on the active and smart packaging, mechanisms of the AM and AMI films, application of the smart films, packaging materials, antimicrobial release mechanism and synergizing of the active and smart film. Chapter 3 provides a detailed methodology of this research to achieve the targeted objectives. Chapter 4 shows the results and discussion on antimicrobial packaging film incorporated with thymol (AM) films. Whilst, chapter 5 presents the results and discussion of the antimicrobial packaging film incorporated with thymol and colour indicators (AMI). Finally, chapter 6 summarises the findings of this study and few suggestions and recommendation for future work.

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