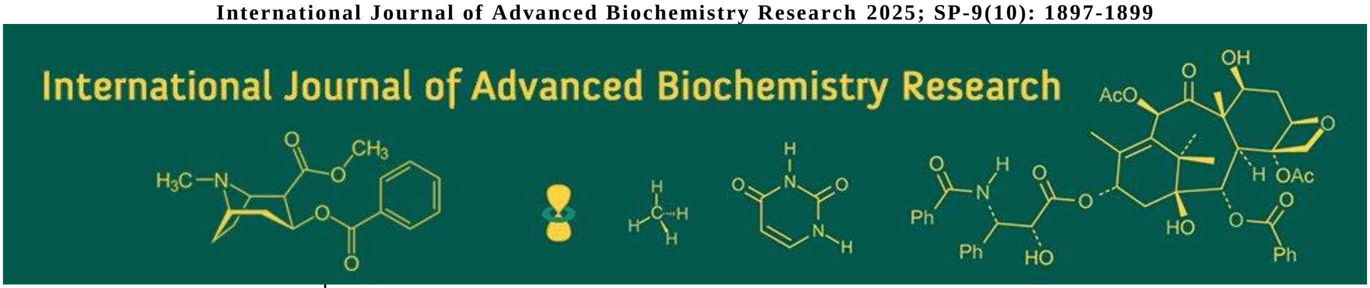

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Development of biodegradable pH-sensitive intelligent films for monitoring freshness of perishable foods

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Abstract
The growing interest in sustainable food packaging has encouraged the development of intelligent films that can signal product freshness through visible color changes. In this study, biodegradable pH-sensitive films were prepared using biopolymers blends (sodium alginate-carboxymethyl cellulose (SA-CMC), sodium alginate-chitosan (SA-CS) and sodium alginate-pectin (SA-P) blends incorporated with anthocyanins extracted from butterfly pea flower (*Clitoria ternatea* L.). Ultrasound-assisted extraction (UAE) of anthocyanins from butterfly pea flower was carried out at 225 W ultrasound power, 35 minutes of sonication time, and a 1:30 g/ml of solid-solvent ratio. Glycerol was used as plasticizer for film development. Results showed that the film formed by using sodium alginate-carboxymethyl cellulose (SA-CMC) blend possess good strength and was the best among three films. It was also observed that the films having glycerol concentration of 30% (w/w, based on total polymer weight) had better flexibility and reduced brittleness. The developed SA-CMC film exhibited clear, uniform coloration and responded to pH changes by shifting from violet to green during chicken storage. Films applied to chicken samples at room and refrigeration temperatures displayed distinct color changes corresponding to spoilage progression. These biodegradable intelligent films show potential as sustainable freshness indicators for perishable food monitoring.

Keywords: Biodegradable film, intelligent packaging, anthocyanin, butterfly pea flower, pH indicator, chicken freshness

1. Introduction

Increasing consumer demand for safe, fresh, and minimally processed foods has accelerated the search for sustainable and intelligent packaging technologies. Conventional plastic packaging, although effective, contributes significantly to environmental pollution and lacks real-time monitoring capabilities. Biopolymer-based films have emerged as eco-friendly alternatives due to their biodegradability, renewability, and film-forming ability (Arrieta *et al.*, 2014; Musso *et al.*, 2019) [1, 7].

Intelligent packaging integrates sensing components to provide information about the condition of the packaged food, such as pH changes, gas evolution, or microbial activity (Yam *et al.*, 2005) [11]. Natural pigments, particularly anthocyanins, are widely explored as pH indicators because of their vivid, reversible color changes under different pH conditions (Khoo *et al.*, 2017) [6]. Butterfly pea flower (*Clitoria ternatea* L.) is a rich source of stable polyacylated anthocyanins that exhibit distinct blue-violet to green transitions across the pH spectrum, making it ideal for developing freshness indicators (Jaafar *et al.*, 2018) [5].

Ultrasound-assisted extraction (UAE) is a green and efficient technique that enhances the release of bioactive compounds by cavitation effects, reducing solvent use and extraction time (Chemat *et al.*, 2017) [2]. Incorporating such extracts into biodegradable matrices like sodium alginate and CMC can produce intelligent films with good mechanical properties and functional sensitivity to spoilage-related pH changes.

This study aims to develop biodegradable intelligent films using biopolymer blends and the extracted pigment from butterfly pea flowers. The developed biodegradable intelligent film was then used as freshness indicator to evaluate their ability to monitor freshness of perishable foods during storage.

2. Materials and Methods

2.1 Materials

Dried butterfly pea flowers (*Clitoria ternatea* L.) were used for anthocyanin extraction. Sodium alginate, carboxymethyl cellulose (CMC), chitosan and pectin served as the main biopolymers. Glycerol was added as a plasticizer. Fresh chicken samples were procured locally. Analytical-grade chemicals were used throughout.

2.2 Ultrasound-assisted extraction of anthocyanins

Extraction was carried out using 5 g of powdered flower (250 µm) with phosphate buffer as solvent maintaining a 1:30 g/ml of solid-solvent ratio. UAE was performed using a probe type ultrasonicator (frequency: 40 kHz; probe diameter: 20 mm) at 225 W ultrasound power for 35 minutes of sonication time. After this, the extracts were filtered, centrifuged, and stored at 4 °C for further use.

2.3 Preparation of intelligent films

1 g of sodium alginate was mixed with 1 g of CMC, chitosan and pectin respectively in 100 ml distilled water at 60 °C under continuous stirring at 500 rpm using magnetic stirrer. After complete dissolution of biopolymers, glycerol was added at a concentration of 30% w/w of the total polymer weight and mixed thoroughly to ensure uniform distribution. After reducing the temperature to 40 °C, flower extract (pigment) was added to the film forming solution. Then, the film-forming solutions were adjusted to pH ≈ 3 for better pigment dispersion, ultrasonicated for 10 min to remove air bubbles, and cast into silicone moulds. Films were dried at 40 °C until uniform sheets were formed.

2.4 Application for chicken freshness monitoring

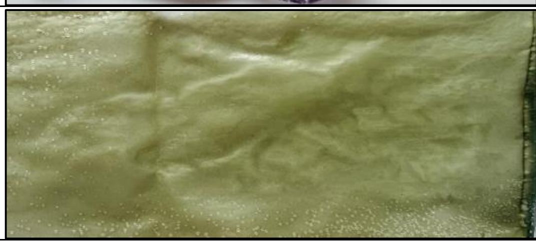
Indicator films were attached inside chicken packaging and stored at room temperature and refrigeration (4 °C). Color changes were monitored visually over time as spoilage progressed, reflecting pH shifts due to microbial activity.

3. Results and Discussion

3.1 Development of biodegradable intelligent films

In the present study, a combination of biopolymers was used for film development instead of a single biopolymer to enhance film properties. It was reported that blending biopolymers improves film flexibility, mechanical strength, and barrier properties compared to films made from a single polymer (Ramesh and Muthukrishnan, 2022) [9]. Thus, in this study, sodium alginate (SA) was selected as the base polymer for the film development due to its excellent film-forming ability, mechanical strength, and biodegradability (Rhim, 2012) [10]. The various biopolymer combinations, including sodium alginate-chitosan (SA-CS), sodium alginate-carboxymethyl cellulose (SA-CMC), and sodium alginate-pectin (SA-P), were tested to determine the best formulation. It was observed that SA-CMC films with 30 % glycerol concentration showed the best flexibility and integrity compared to alginate-pectin or alginate-chitosan blends. Among the combinations tested, SA-CMC films exhibited superior uniformity, structural integrity, and compatibility with butterfly pea flower extract. These observations align with reports that blending alginate with CMC improves film homogeneity and barrier properties (García *et al.*, 2014) [4]. Table 1 shows the findings of various films combinations.

Table 1: Development of biodegradable intelligent films using different biopolymers blends

Film combinations	Findings	
SA-CS-BPFE	Yellow colored film was developed, not retaining desired color of pigment, difficulty in film forming, brittle	
SA-P-BPFE	Purple colored film, more flexible (crumbled), less strength, stickiness	
SA-CMC-BPFE	Light greenish film, good strength, no stickiness, best among three films	

SA: Sodium alginate, CS: Chitosan, P: Pectin, CMC: Carboxymethyl cellulose, BPFE: Butterfly pea flower extract

3.3 Freshness Monitoring Application

The developed intelligent film was used as freshness indicator to monitor the freshness of perishable foods. SA-CMC films were cut into square shaped labels and attached to the inside lid of the sterile air-tight box. Fresh raw chicken was kept inside the boxes and stored at room temperature and under refrigeration respectively. It was observed that the films exhibited visible color changes when attached to chicken packaging (Fig. 1). At room temperature, color shifted from violet to green within a shorter period due to rapid spoilage, while at 4 °C, the transition occurred more slowly. This corresponds to increasing volatile amines and pH during storage. Similar anthocyanin-based indicators have effectively tracked fish and meat freshness through color shifts (Puligundla *et al.*, 2022) [8].

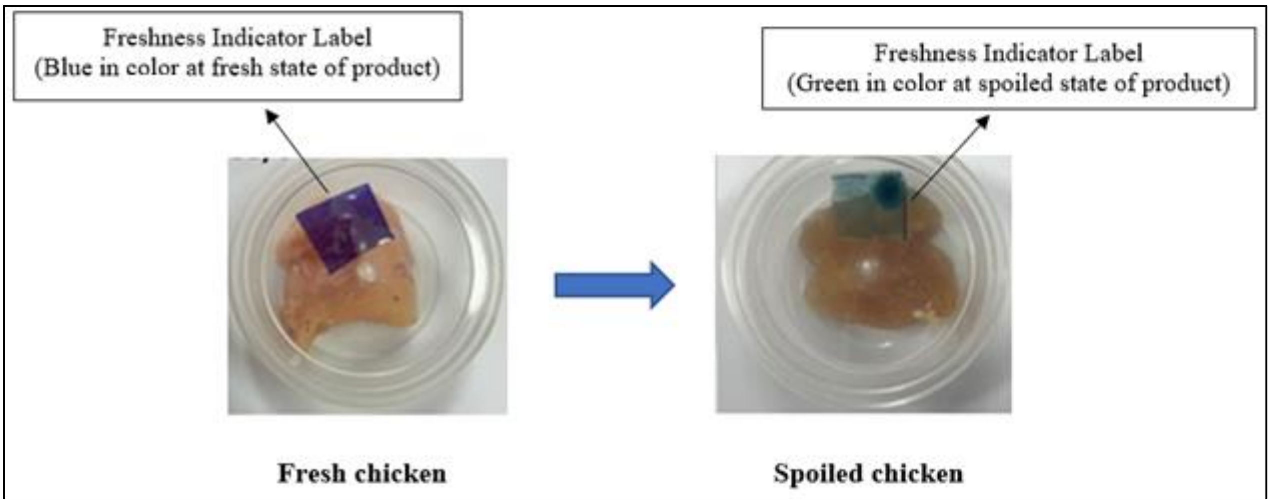


Fig 1: Visible color changes in the intelligent film during storage of chicken

The results confirm that the developed films are sensitive to environmental pH changes associated with microbial spoilage, making them practical for real-time freshness monitoring.

4. Conclusion

Biodegradable pH-sensitive intelligent films were successfully developed using biopolymer blends incorporated with anthocyanins from butterfly pea flower extracted via UAE. SA-CMC blend was found to be the best among other film combinations that have better appearance, film flexibility and good strength. The best developed film formulation showed good film-forming properties and responded distinctly to spoilage-related pH changes in chicken stored at different temperatures. These films offer a promising, eco-friendly approach to intelligent food packaging for real-time freshness indication.

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