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Chromaticity and color saturation of ultraviolet irradiated poly(vinyl alcohol)-anthocyanin coatings

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ABSTRACT

The purpose of this paper is to evaluate the chromaticity and color saturation of anthocyanin extraction from fruit pericarps of *Ixora siamensis* in a poly(vinyl alcohol) (PVA) matrix. The colored PVA matrix was exposed to UV-B irradiation for 93 days at UV intensity of 17.55 lux. Anthocyanin colorant has been extracted using methanol acidified with 0.5% trifluoroacetic acid (TFA). Different concentrations of ferulic acid (FA) (0, 1, 2, 3, 4 and 5 wt.%) have been added to the anthocyanin extractions before mixing with PVA to form a coating system. The PVA-anthocyanin-FA mixtures have been coated on glass slides and kept overnight in the dark for curing before exposure to UV-B irradiation. The FA-free sample undergoes more color degradation compared to samples containing FA. The coating with 2% FA has the most stable color with chromaticity of 41% and color saturation of 0.88 compared to other FA containing coats. The FA-free coat exhibits 29% chromaticity and color saturation of 0.38 at the end of the experiment.

1. Introduction

Pigments absorb light in the visible and their colors depend on the chromophores present [1]. Natural pigments are less stable compared to synthetic dyes or colorants, but have attracted much attention in the food and pharmaceutical industries. Anthocyanins are responsible for the different colorations in plants. The range of colors depends on the degree of oxygenation and the number of sugar moieties in anthocyanidin [2]. Anthocyanidin is the central chromophore for anthocyanin. The basic structure of anthocyanin is shown in Fig. 1 [3].

Anthocyanin color degradation can be delayed with co-pigmentation. Co-pigmentation can result in anthocyanin absorption to be red-shifted with an increase in absorption and stability [4]. Co-pigmentation of anthocyanins with flavonoids, alkaloids, amino acids, organic acids, nucleotides, polysaccharides, metals or other anthocyanin itself [5] is the main mechanism of color stabilization in plants [6,7]. Abyari et al. [8], Setareh et al. [9] and Yawadio and Morita [10] have used co-pigmentation to delay color degradation.

In this work, ferulic acid (FA), the structure of which is shown in Fig. 2, has been chosen as a co-pigment to delay color degradation of anthocyanin extracted from the fruit pulp of *Ixora siamensis*. The phenolic nucleus of FA has an extended side chain conjugated in its structure. This can form a resonance stabilized phenoxy radical, which neutralizes damage causing free radicals [11]. Due to these properties, FA is chosen as the co-pigment.

Color is one of the most important factors that defines the quality of a product and has a decisive influence on the acceptance or rejection of a product by consumers. The effects of co-pigmentation on the color of the coatings before and after exposure to UV-B irradiation (with intensity 17.55 lux) for 93 days have been analyzed using the Commission Internationale de l'Eclairage (CIE) color system.

2. Experimental setup

2.1. Materials

The fruits of *I. siamensis* were collected in Banting, in the state of Selangor, Malaysia where they are locally known as "*jejarum*". The fruits were sealed in polyethylene bags, covered with aluminum foil and kept frozen at (-18 ± 2) °C before being processed. A 300 g mass of *I. siamensis* fruit pericarps was ground using a mortar and pestle. Anthocyanin was extracted using methanol containing 0.5% trifluoroacetic acid (TFA) (v/v). The anthocyanin extract was centrifuged at 10,000 rpm for 15 min. The supernatant liquid was then filtered using Whatman No. 1 filter paper to remove residues. The methanol was then evaporated under reduced pressure at

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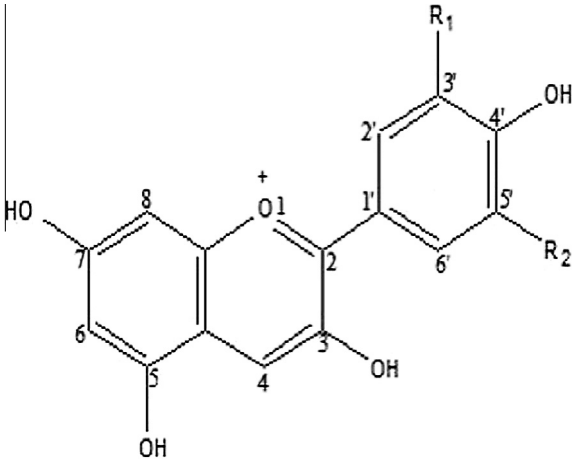

Fig. 1. Basic structure of anthocyanin.

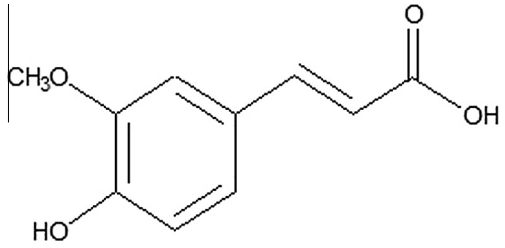

Fig. 2. Structure of ferulic acid.

room temperature. Five different concentrations of ferulic acid (FA) co-pigment (0, 1, 2, 3, 4 and 5 wt.%) were added to improve the color stability of the anthocyanin. These solutions were then mixed with PVA and "painted" onto a glass slide to form a coating system. Every 1 g of coating mixture contains 70% anthocyanin and 30% PVA by weight. The coats were kept overnight in the dark for curing process. All samples were prepared in triplicate. The color of the samples was then subjected to the CIE color analysis [12]. From this analysis, the most stable color in terms of FA content was obtained.

2.2. Spectrophotometric measurements

2.2.1. Color analysis determination

The PVA-anthocyanin-FA coatings were placed under UV-B lamp in a dark room. The distance between the samples and the light source was fixed at 5 cm. All prepared coatings were exposed to UV-B irradiation (312 nm) at 17.55 lux intensity for 93 days. Spectral curves were also recorded in dark with a regular transmission, from 380 to 780 nm using the Avantes (AvaSoft 7.6) visible spectrophotometer and analyzed using the CIE color system.

3. Results and discussion

3.1. CIE chromaticity (C^*) for PVA-anthocyanin blends

The chromaticity (C^*) values at different times from the beginning until the end of exposure for PVA-anthocyanin blends are shown in Fig. 3. Chromaticity (C^*) corresponds to color brightness and is generally observed by the color intensity [13]. The chromaticity C^* of FA-free sample at the beginning of exposure was 38.70 ± 0.01 and it decreased to 29.17 ± 0.02 at the end of 93 days

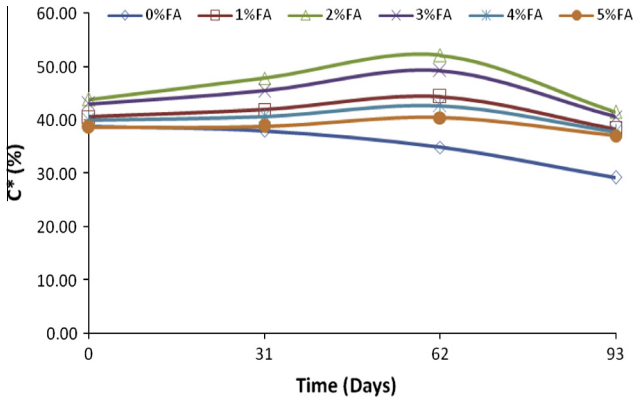

Fig. 3. C^* values (%) for PVA-anthocyanin blends during 93 days of exposure for coats of different FA concentrations.

exposure, indicating a 24% difference in chromaticity. According to Tsurunaga et al. [14] the UV-B irradiation with wavelength greater than 300 nm increased anthocyanin levels. However, our result implied otherwise. Buckwheat sprouts has been reported to accumulate anthocyanin glucoside and rutinoside for protection against UV stress in the presence of sunlight. These reports indicated that sunlight and ultraviolet irradiation increased the anthocyanin content; hence carrying out the investigation in the dark can result in significantly lower anthocyanin content [15].

On the other hand, C^* values for PVA-anthocyanin blends with added FA increased from time zero until 62nd day of exposure before decreasing until the end of the experiment.

The PVA-anthocyanin blends with 2 wt.% added FA exhibited the highest C^* (brightest color). At time zero, $C^* = 43.74 \pm 0.01$, which increased to 47.72 ± 0.01 on the 31st day of exposure and then to 52.03 ± 0.01 on the 62nd day. After the 62nd day C^* decreased to 41.39 ± 0.01, but is still higher than that of the FA-free coating. The color intensity of samples with 2 wt.% FA showed a slight drop of ~5% from the initial value, implying the better performance of the coating.

These findings agreed with studies by Bakhshayeshi et al. [16] and Janna et al. [17] who showed that UV irradiation leads to destruction and bleaching of color for untreated anthocyanin. Bakhshayeshi et al. [16] and Janna et al. [17] studied the influence of UV on anthocyanin from the species of *Malus* varieties and *Tibouchina semidecandra* L., respectively. In these works, the color of the samples with 2 wt.% FA resulted in the brightest color with the highest C^*. Bakowska et al. [18] also reported that the presence of flavones, tannic and chlorogenic acids in *Lonicera kamtschatica* anthocyanin will inhibit degradation by UV.

FA helped dissipate the absorbed UV energy and prevented color degradation in PVA-anthocyanin coat. The strong capability of FA as UV absorber is due to its phenolic nucleus and extended side chain conjugation, which readily forms a resonance that stabilizes phenoxy radical. The results of this study show that the addition of FA enhanced color brightness (C^*) throughout the 93 days of exposure. However, at higher concentrations of FA, from 3 to 5 wt.%, the co-pigmentation showed reduced color brightness (C^*) compared to the 2 wt.% FA added sample. This observation is supported by Sun et al. [19] who studied the effects of different co-pigments such as sinapic, ferulic, caffeic, coumaric and gallic acids on *Rubus idaeus* L., *Heritage* anthocyanin color stability. They found that when the co-pigment concentration exceeds a certain level, no further enhancement in color properties was observed. Therefore, the molar ratio cannot be raised indefinitely. This explains why the addition of 3–5 wt.% FA did not further enhance