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Curcumin and PLA-Curcumin Coating's Chromaticity and Colour Stability in Reaction to the Effectiveness of pH

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ABSTRACT

Understanding the limitations and unfavourable conditions of curcumin would be an advantage to increase the stability of the pigment. Therefore, the ultimate objective of this research is to evaluate the effects of pH on chromaticity and stability of curcumin and PLA-curcumin extracted from *Curcuma longa* (turmeric), as a potential natural source for coating. Overall, the dark condition was preferable and stable for the curcumin brightness and slower the degradation percentage. Dark condition exhibited a better and stable colour degradation (approximately 40%) in all curcumin concentrations (1 mg/ml, 2 mg/ml and 3 mg/ml) as compared to light condition with 89.92%, 85.79% and 60.90%, respectively. PLA alone showed the highest chroma (C^*) value at 3.08% in dark condition, which was slightly higher than in light condition (2.87%). The highest C^* values were achieved in curcumin set with lower pH (pH 3, 5 and 7) and the lowest at higher pH (pH 9 and 11) in both light and dark conditions. However, all PLA-curcumin showed colour stability in both acidic (pH 3 and 5) and basic (pH 9 and 11) in dark condition only.

Keywords: Curcumin, PLA-curcumin, pH, Chromaticity, Colour stability, Natural pigment

1 Introduction

Most carotenoids are known as lipid-soluble pigments. However, curcumin is a water-soluble carotenoid that generally extracted with water or lower alcohols. The identity of individual carotenoids was confirmed by their spectral characteristics, absorption maximum and retention time [1]. According to [2], the detection of individual carotenoids was made at the wavelengths of maximum absorption of the carotenoids in the mobile phase: neoxanthin (438 nm), violaxanthin (441 nm), lutein (447 nm) zeaxanthin (452 nm), β -carotene (454 nm), β -cryptoxanthin (450 nm) and α -carotene (456 nm). [2] reported on the single peak of curcumin (extracted from turmeric/*Curcuma longa*) that was gained from HPLC analysis at 425 nm with 12.95 ± 1.07 mg/g DW (the highest curcumin value as compared to *Curcuma xanthorrhiza*, *Curcuma manga*, *Zingiber zerumbet* and *Zingiber cassumunar*. Furthermore, carotenoids



have attracted greater attention due to their beneficial effect on human health such as involvement in cancer prevention, reduction of the risk for degenerative diseases such as cardiovascular diseases, macular degeneration and cataract and also enhancement of immune response [3].

Polymers are substances with a long chain of repetitive smaller units bonded together. It can be classified into two categories based on their origins: petrochemical-based or natural-based [4] and have a wide range of applications from plastics as packaging materials. However, these convenient petrochemical-based materials that are non-biodegradable and designed for immediate disposal have caused pollution problems ever since they became dominant in the marketplace around 1940 [5]. Fortunately, Poly (lactic acid) or PLA, is a green polymer from renewable resources (manufactured by the polymerization of lactic acid monomers derived from the fermentation of starch feedstocks) that were biodegradable and compostable (bioplastic). However, it has lower strength, brittle, flammable and high permeability to most gases and solvents as compared to metal and ceramics, which limits its application in the industry [6]. Understanding the limitations and unfavourable conditions of curcumin would be an advantage to increase the stability of the pigment. Therefore, the ultimate objective of this research is to evaluate the effects of pH on chromaticity and stability of curcumin and PLA-curcumin extracted from *Curcuma longa* (turmeric), as a potential natural source for coating.

2 Materials and Methods

2.1 Chemical Extraction of Curcumin

The curcumin extraction followed the methods described by [7] and [8; 9] all parts of turmeric (*Curcuma longa*) rhizome samples were freeze-dried for 72 h, before grounded into a fine powder and kept at -20°C. The powdered sample (0.1 g) was rehydrated with distilled water and extracted with a mixture of acetone and methanol (7:3) at room temperature until it turned colourless. The crude extract was centrifuged for 5 min at 10,000 rpm and stored at 4°C in the dark prior to analysis. To extract the carotenoid (curcumin), an equal volume of hexane and distilled water were added to the combined supernatants. The solution was then allowed to separate and the upper layer containing the carotenoids was collected. The combined upper phase was then dried to completion under a gentle stream of oxygen-free nitrogen.

2.2 Spectrophotometric measurements

The yellow colour intensity of curcumin was measured by using a Remote DRA Cary-50 chromameter. The analysis abbreviated as CIELAB refers to the Commission Internationale de l'Éclairage (International Commission on Illumination) colour system, which contains lightness (L^*) and colours (a^* and b^*): L^* = lightness, a^* = red-purple (+)/bluish-green (-) hue component, b^* = yellow (+)/blue (-) hue component, C^* [$(a^{*2} + b^{*2})^{1/2}$] = chroma, h° (from arctangent b^*/a^*) = hue angle (0° = red-purple, 90° = yellow, 180° = bluish-green, 270° = blue).

The L^* value ranges from black (0) to white (100), positive a^* indicates a hue of red-purple and negative a^* , of bluish-green. Whereas positive b^* indicates yellow and negative b^* blue. The colour of achromatic or neutral grey is when the point a^* and b^* axes cross, at the L^* value of 50.

CIELAB colour parameters (a^* , b^* , L^* and ΔE^*_{Lab}) were analysed using UV-Vis spectrophotometer (Shimadzu UV-160 Spectrophotometer) by adopting the method of [6] with some modifications. In order to calculate the colour changes, the differences in chromatic coordinates (Δa^* and Δb^*), lightness (ΔL^*) and the total colour difference (ΔE^*_{Lab}) were calculated using the following equations: $\Delta a^* = a^* - a_0^*$; $\Delta b^* = b^* - b_0^*$; $\Delta L^* = L^* - L_0^*$; $\Delta E^*_{Lab} = (\Delta a^{*2} + \Delta b^{*2} + \Delta L^{*2})^{1/2}$. Where a_0^* , b_0^* and L_0^* are the initial values before each treatment. The values of each colour parameter are expressed as the mean of three determinations. The relative standard deviation (RSD) was less than 0.2% for any measurement, indicating a high reproducibility [7].

2.3 Effects of pH

Five different pH (pH 3, 5, 7, 9 and 11) ranging from acidic to basic and three different curcumin concentrations (1 mg/mL, 2 mg/mL and 3 mg/mL) were dissolved in EtOH. The pH of the solution was adjusted by adding either drop of 10% NaOH solution to make it basic, or 10% HCl solution to make it acidic. The samples were placed in both dark and light conditions. Each of the samples was prepared in triplicate. The absorbance readings using a Remote DRA Cary-50 chroma meter were taken weekly, for four weeks.

2.4 PLA-curcumin

PLA beads were initially dried in the oven at 65°C for 15 minutes to remove any remaining moisture. The dried PLA beads were weighed (5 g) in a beaker and added with Chloroform (50 mL). The mixture was stirred using a magnetic stirrer until the PLA beads were dissolved completely. Then, curcumin solution (that dissolved in EtOH) was added. The solution (5 mL) was pipetted into a 65 mm diameter glassed petri dish. The petri dish was placed under the fume chamber for 24 h until the mixture dried up.

The parameters used were the same as the curcumin stability test. The treatment to the polymer was done inside the petri dish by adding a solution of intended pH into the polymer. Then, the petri dish was left for about 30 minutes. The polymer was then dried from an excess of the treatment solution by removing it and dabbing the polymer with tissue paper. The duration of the experiment was set for 4 weeks. The absorbance readings were taken using a Remote DRA Cary-50 chromameter.

3 Results

The CIE $L^*a^*b^*$ color difference (ΔE^*) is used to assess the stabilization of pigment particles. If ΔE^* value < 1.0, it is an indication that no colour difference is visible, if ΔE^* value < 2.0,

indicate the colour difference is only slightly visible, ΔE^* value < 3.5 indicate noticeable colour difference, ΔE^* value < 5.0 indicate clear difference in colour is noticed whereas ΔE^* value > 5.0 indicate clearly two different colours [10]. Chromaticity or colour difference relative measurements of curcumin pigment from 1 mg/mL up to 3 mg/mL as well as the time of exposure from week 0 until week 4. There is a strong relationship between the intensity of the curcumin yellow colour concentration and period of time in response to light and dark condition. This data suggests that although different curcumin concentrations had influenced yellow intensity, light and period of time also had an effect on yellow intensity and stability.

4 Discussion

In this research, the results revealed the ability of pH to influenced the activities of curcumin and PLA-curcumin. Overall, the dark condition was preferable and stable for the curcumin brightness and slower the degradation percentage (approximately 45-60% better than light condition). The C^* values showed a maximum in PLA-curcumin as compared to curcumin alone. Therefore, the presence of PLA in curcumin will stabilized the chromaticity (brightness) and delayed the colour degradation. Therefore, PLA-curcumin is potential as non-toxic and biodegradable polymer especially for disposable packaging (bioplastic) and coating industry. There are many different types of polymers that can be used, including bioplastics, biopolymers, and commercial plastics. Biopolymers are one of these and are used to draw attention due to their reproducibility, biodegradability, and excellent functional properties [11]. Curcumin may also be thought of as immobilised in a film matrix, which demonstrated a pronounced pH-responsive colour change needed for the applications of intelligent food packaging. Due to the alteration in its dominant structure, which is studied under various pH conditions, curcumin can change colour, which can be used to monitor food quality in real-time [12]. Since curcumin has pH-dependent colour change properties and functional properties that can be used in food packaging applications, it has attracted the attention of numerous scientists in recent years. This is because curcumin may help increase the food's quality and shelf life.

5 Conclusion

Natural colourants derived from curcumin are easily subject to deterioration and susceptible to environmental factors, such as pH, which will restrict their use and commercial viability. As a result, research must be done to assess the natural colorant's colour properties, particularly during exposure time, and based on the findings, methods will be developed to enhance and improve the properties. To assess the colour stability of curcumin on coating systems, CIE colour analysis was used in this study. Product colour is a crucial quality attribute. The CIE parameter can be used to measure colour and track colourant degradation. This is so that it can be applied to all colours that the human eye can perceive. By analysing

the colours in terms of E and s and using the CIE parameter ($L^* C^* H^\circ a^* b^*$), the colours can be accurately described.

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Competing Interests

The authors declare no conflict of interest. The funders had no role in the design of the study; in the collection, analyses, or interpretation of data; in the writing of the manuscript; or in the decision to publish the results.

References

1. Britton, G., Liaaen-Jensen, S. and Pfander, H. "Carotenoids. Vol. 1A: Isolation and analysis", Birkhauser Verlag, Boston, 1995.
2. Jambeck, J., Geyer, R., Wilcox, C. "Marine pollution. Plastic waste inputs from land into the ocean", *PubMed* 2015, 347(6223), pp. 768-771.
3. Mochizuki, K. and Takayama, K. "Prediction of color changes in acetaminophen solution using the time-temperature superposition principle", *Drug Development and Industrial Pharmacy* 2016 42(7), pp. 1050-7.
4. Williams, C.K. and Hillmyer, M.A. "Polymers from renewable resources: A perspective for a special issue of polymer reviews", *Polymer Reviews* 2008, 48, pp. 1-10.
5. Jambeck, J., Geyer, R., Wilcox, C. "Marine pollution. Plastic waste inputs from land into the ocean", *PubMed* 2015, 347(6223), pp. 768-771.
6. Shirai, M., Grossmann, M., Mali, S., Yamashita, F., Garcia, P. and Müller, C. "Development of biodegradable flexible films of starch and poly (lactic acid) plasticized with adipate or citrate esters", *Carbohydrate Polymers* 2013, 92(1), pp. 19-22.
7. Othman, R. (2009), "Biochemistry and genetics of carotenoid composition in potato tubers", Christchurch, New Zealand: Lincoln University, PhD thesis.
8. Othman, R., Noh, N.H., Ahmad, N., Anis, H. and Jamaludin, M.A. "Determination of natural carotenoid pigments from freshwater green algae as potential halal colorants", *International Food Research Journal* 2017, 24, pp. S468-S471.
9. Othman, R., Abdurasid, M., Mahmad, N. and Ahmad Fadzillah, N. "Alkaline-based curcumin extraction from selected zingiberaceae for antimicrobial and antioxidant activities", *Pigment & Resin Technology* 2019, 48(4), pp. 293-300.
10. Mokrzycki, W. and Tatol, M. "Color difference Delta E - A survey", *Machine Graphics and Vision* 2011, 20(4), pp. 383-411.
11. Rouf, T. B., and Kokini, J. L. (2018), Natural Biopolymer-Based Nanocomposite Films for Packaging Applications, In: Jawaid, M., Swain, S. (eds), *Bionanocomposites for Packaging Applications*, Cham: Springer International Publishing, pp. 149-177. https://doi.org/10.1007/978-3-319-67319-6_8.
12. Priyadarshi, R., Roy, S., Ghosh, T., Biswas, D., & Rhim, J.-W. "Antimicrobial nanofillers reinforced biopolymer composite films for active food packaging applications - a review", *Sustainable Materials and Technologies* 2021, 32. <https://doi.org/10.1016/j.susmat.2021.e00353>