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EFFECT OF pH AND UV EXPOSURE ON THE CHROMATICITY STABILITY OF CURCUMIN EXTRACTED FROM TEMU LAWAK (*Curcuma xanthorrhiza* Roxb.)

(KESAN pH DAN PENDEDAHAN UV TERHADAP KESTABILAN KROMATIK EKSTRAK KURKUMIN DARIPADA TEMULAWAK (Curcuma xanthorrhiza ROXB.)

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Abstract

The traditional uses of the family *Zingiberaceae* have a long history and include everything from folk medicine to culinary applications. Numerous ginger species have been the subject of numerous phytochemical, pharmacological, and molecular studies worldwide. Therefore, this study was implemented to explore and investigate in greater depth of curcumin extracted from ginger species in a wide range of rhizome flesh colour. UV-Vis spectrophotometer and CIELAB chromameter were used to measure the curcumin chromaticity stability. The chromaticity stability of curcumin extracted from *temu lawak* (*Curcuma xanthorrhiza* Roxb.), as analyzed using the CIELAB colour system, was found to be more stable at lower pH levels (3, 5, and 7) compared to higher pH levels (9 and 11), indicating that acidic to neutral conditions help preserve the colour integrity of curcumin. In addition the curcumin pigment was more stable in UV-B at higher concentrations as compared to UV-A.

Keywords: Curcumin, chromaticity, carotenoid, natural pigment, temu lawak

Abstrak

Kegunaan tradisional keluarga Zingiberaceae telah lama wujud dan merangkumi pelbagai aspek daripada perubatan tradisional hingga ke aplikasi kulinari. Pelbagai spesies halia telah menjadi subjek kajian fitokimia, farmakologi dan molekul di seluruh dunia. Oleh itu, kajian ini dilaksanakan untuk meneroka dan menyelidik dengan lebih mendalam tentang kurkumin yang diekstrak daripada spesies halia dengan pelbagai warna isi rizom. Spektrofotometer UV-Vis dan kromameter CIELAB digunakan untuk mengukur kestabilan kromatik kurkumin. Kestabilan kromatik kurkumin yang diekstrak daripada temu lawak (Curcuma xanthorrhiza Roxb.), seperti yang dianalisis menggunakan sistem warna CIELAB, didapati lebih stabil pada tahap pH yang rendah (3, 5 dan 7) berbanding tahap pH yang tinggi (9 dan 11). Ini menunjukkan bahawa keadaan berasid hingga neutral membantu mengekalkan integriti warna kurkumin. Selain itu, pigmen kurkumin lebih stabil di bawah sinaran UV-B pada kepekatan yang lebih tinggi berbanding UV-A.

Kata Kunci: *Kurkumin, Kromatisiti, karotenoid, pewarna alami, temu lawak*

INTRODUCTION

In early human history, the exclusive use of natural materials was not a matter of choice but rather a necessity, as synthetic or artificial alternatives had yet to be developed. Organically sourced materials were often preferred due to their superior quality compared to manufactured substitutes. Colour has always played a vital role in shaping human cultures across the world. Over time, public awareness has grown concerning environmental preservation, health safety, and the sustainability of non-toxic, naturally derived colourants. These eco-friendly alternatives are increasingly favoured due to their minimal environmental impact. The perception of colour arises from the emission of specific frequencies within the electromagnetic spectrum by light sources (Delgado-Vargas & Paredes-López 2003).

The U.S. Food and Drug Administration (FDA) defines a colour additive as any material, unless exempt under section 201(t) of the Act, that contributes to dyes, pigments, or other substances produced through synthesis or similar processes. This includes materials that are extracted, isolated, or derived either as intermediates or final products from vegetable, mineral, animal, or other sources and used in food, drugs, cosmetics, or on the human body to impart colour (CFR 2016a). Beyond its technical scope, this definition reflects a fundamental human tendency (fitrah) towards appreciating beauty, vibrant hues, and aesthetic detail an inclination deeply rooted in cultural expressions. The emergence of synthetic dyes in the late 1800s marked a turning point in colourant use, offering a wide range of applications (Ul-Islam 2017). Today, consumers are increasingly concerned about the safety of synthetic colourants. Their widespread use is largely due to economic benefits, including low cost, high stability, and resistance to environmental factors such as light, oxygen, and pH variations (Sharma et al. 2021). Synthetic dyes are also more affordable and can be produced in larger volumes than natural alternatives (Mittal 2020a).

However, the supply of natural colourants is often constrained by seasonal and resource limitations (Mittal 2020b). Despite their practical advantages such as consistency, cost-effectiveness, and strong colouring properties synthetic colourants pose significant health risks when consumed over time. These risks include anaemia, urticaria, kidney damage, indigestion, allergic reactions, and the development of tumours or cancerous lesions in vital organs such as the brain, spleen, and liver. Other potential effects include birth defects, mental and growth retardation, and vision problems that may lead to blindness (Sharma et al. 2021). Consequently, artificial food colours have long been the subject of public and scientific scrutiny due to their association with allergies, hyperactivity, and carcinogenicity (Sharma et al. 2021).

Temulawak is a medicinal plant from the Zingiberaceae family. At present, most temulawak cultivated in Indonesia, Malaysia, Thailand, and Philippines, where it thrives in lowlands areas at altitudes of 1500 m above sea level and in tropical forests (Kustina et al. 2020). The use of temulawak as a traditional medicine has been increasing at an average of 5.4% annually over the last decade (Nihayati et al. 2013), This has been triggered by the rise in public awareness and consumption of alternative therapeutic treatments with fewer side effects. Due to the rising prices of chemical medications more than what societies can afford, people have been turning to natural sources for healing (Sumarya et al. 2020).

The rhizome of the curcuma plant is nutritious as it contains chemical compounds, such as curcumin, essential oils, saponins, flavonoids, alkaloids, and tannins. It is traditionally used to treat heartburn, diarrhoea, piles, cough, asthma, and canker sores. The properties of temulawak include increasing appetite, improving digestive function, maintaining, and improving liver function, relieving joint and bone pain, reducing blood and fat concentrations in the body, and serving as an antioxidant (Nihayati et al. 2013). As seen in Figure 1, curcumin is a yellow pigment that has been extracted from the rhizome of the curcuma plant. Curcuma very closely correlates with many bioactive compounds, especially curcuminoids, which are a group of phenolic compounds that comprise curcumin, demethoxycurcumin, and bisdemethoxycurcumin (Sari et al. 2013; Anggoro et al. 2015).

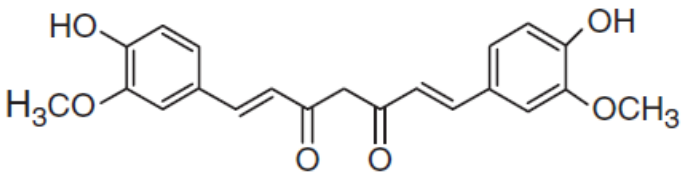

Figure1. Chemical structure of curcumin

Due to its properties as a safe, natural colorant and its numerous health benefits, *Curcuma xanthorrhiza* (temulawak) presents significant potential for large-scale cultivation and commercial application in Malaysia. The rising consumer demand for natural and Halal-certified products further supports the relevance of temulawak in the food, pharmaceutical, and cosmetic industries. As consumers become increasingly conscious of product ingredients and their sources, the use of plant-based additives like curcumin aligns well with current market trends and ethical expectations, especially within the Halal industry. Moreover, curcumin, the main pigment compound found in temulawak, has demonstrated antioxidant, anti-inflammatory, and antimicrobial properties, making it highly valuable not only as a colorant but also as a functional ingredient.

However, despite its promising applications, the stability of curcumin remains a major challenge, especially under certain environmental conditions such as high pH, elevated temperatures, and light exposure. These factors can lead to rapid degradation, which limits its effectiveness and shelf life in various formulations. Therefore, a deeper understanding of the limitations and unfavorable conditions affecting curcumin stability is essential. Optimizing formulation methods, storage conditions, and possibly applying encapsulation technologies could significantly enhance the stability and broaden the application of curcumin as a natural and Halal-compliant colorant in diverse industries.

MATERIALS AND METHODS

CURCUMIN STABILITY TEST

Pigments preparation

The maximum concentration of curcumin obtained by chemical extraction and stored at -20 °C was used for colour stability tests. Curcumin has been dissolved in ethanol and distilled H_2O to yield three different concentrations: 1, 2, 3 g/L, respectively. The chromaticity of the untreated sample (ΔE*) was evaluated as a blank.

Analysis of Chromaticity

A chromameter was used to measure the colour intensity of the extracted curcumin. The CIELAB colour system comprises L*, a*, b*, C*, and h°, with L* ranging from black (0) to white (100), and where a positive a* indicates a hue of red-purple, a negative a* indicates a hue of bluish-green, a positive b* indicates a hue of yellow and a negative b* indicates a hue of blue. The colour is considered achromatic or neutral grey when the points of the a* and b* axes cross and L* = 50. The CIELAB colour parameters, namely, a*, b*, L*, and ΔE*Lab, were analysed using a Mettler Toledo® UV7 UV-Vis spectrophotometer according to the method proposed by Mochizuki and Takayama (2016). The used of CIELAB system for analysing L*, a*, and b* values of all samples (Figure 2).

Equation (Yawadio and Morita 2007; Licȯn et al. 2012) was used to calculate the 3 measured colour factors where C* represents colour intensity and brightness, h° indicates the colour shade, and ΔE (Licȯn et al. 2012; Yawadio and Morita 2007).

$$\Delta E = [(\Delta L^*)2 + (\Delta a^*)2 + (\Delta b^*)2]1/2$$

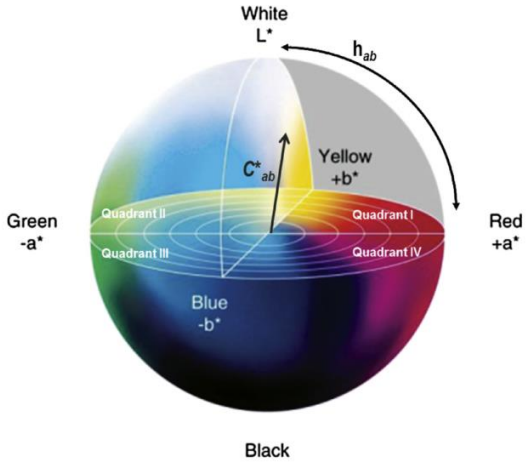

Figure 2. CIELab sphere (Sant' Anna et al. 2013

pH Test

The pH of the samples was adjusted to 3, 5, 7, 9, and 11 using hydrochloric acid and sodium hydroxide. Samples were prepared at three different concentrations and exposed to both dark and light conditions (cool light from a fluorescent lamp with an intensity of 3100 lux, positioned perpendicularly 30 cm below) at room temperature for four weeks. The samples were prepared in triplicate, with chromaticity monitored hourly. Additionally, the chromaticity of the samples was measured on the first day.

UV Irradiation Test

All glass tubes containing samples at three different concentrations were placed in a laminar hood and exposed to UV-A (long wavelength = 365 nm, intensity = 5500 lux) and UV-B (middle wavelength = 312 nm, intensity = 2900 lux) perpendicularly at 10 cm from the light source for 8 hours at room temperature. The samples were prepared in triplicate, and their chromaticity was analysed hourly to monitor changes. Any colour alterations before and after the experiment were measured accordingly.

RESULTS

Effect of pH

An analysis was conducted to examine the correlation between the ΔE*Lab, s, and b* for different concentrations of curcumin in light and dark conditions for different exposure times over a range of pH (3, 5, 7, 9, and 11). Figure 5.6 shows that the ΔE* was > 5.0 for all three concentrations of curcumin pigment after treatment with solutions with pH values of 3, 5, 7, 9, and 11 in both light and dark conditions, except for the curcumin at a concentration of 1 mg/ml for the first week of exposure in solutions with pH values of 3, 5 and 7. The curcumin pigments at pH 9 and pH 11 were found to degrade at a faster rate compared to at pH 7, and the degradation was lower when the ΔE* was > 10.0. The results depicted in Table 1, Figures 4 and 5 show that increasing the pH, curcumin concentration, time of exposure and light caused greater degradation of s and b* in the curcumin pigment. The degree of degradation of the s and b* of the curcumin at pH 3, 5 and 7 was between 22.80% to 39.50% and 22.48% to 51.86% in light conditions, and 20.79% to 32.92% and 22.63% to 40.84% in dark conditions, respectively. This was considerably lower than the degradation of the s and b* of the curcumin at pH 9 and 11 of 35.12% to 85.09% and 36.05% to 94.54% in light conditions, 37.56% to 90.47% and 42.93% to 96.40% in dark conditions, respectively.

Table 1. The effects of pHs of 3, 5, 7, 9 and 11 on the colour degradation of curcumin pigment concentrations of 1, 2 and 3g/L extracted from *temulawak* and exposed to dark and light conditions post-four weeks.

		Degradation percentage (%)					
		Light			**Dark**		
		Curcumin concentration (g/L)					
CIE L*a*b*	**pH**	**1**	**2**	**3**	**1**	**2**	**3**
Colour saturation (s)	3	22.80	30.01	33.23	**20.79**	**25.22**	**32.14**
	5	23.00	30.05	33.48	**20.91**	**31.87**	**32.50**
	7	24.19	39.15	39.50	**20.31**	**37.12**	**32.92**
	9	35.12	44.02	43.40	**37.56**	**50.26**	**47.14**
	11	64.48	80.25	85.09	**65.67**	**84.04**	**90.47**
Yellow intensity (b*)	3	22.48	31.75	40.72	**22.63**	**31.67**	**41.65**
	5	22.63	31.67	41.65	**19.81**	**32.26**	**39.70**
	7	24.32	41.46	51.86	**19.66**	**37.59**	**40.84**
	9	36.05	54.11	58.47	**42.93**	**66.13**	**73.95**
	11	83.91	92.92	94.54	**87.92**	**95.02**	**96.40**

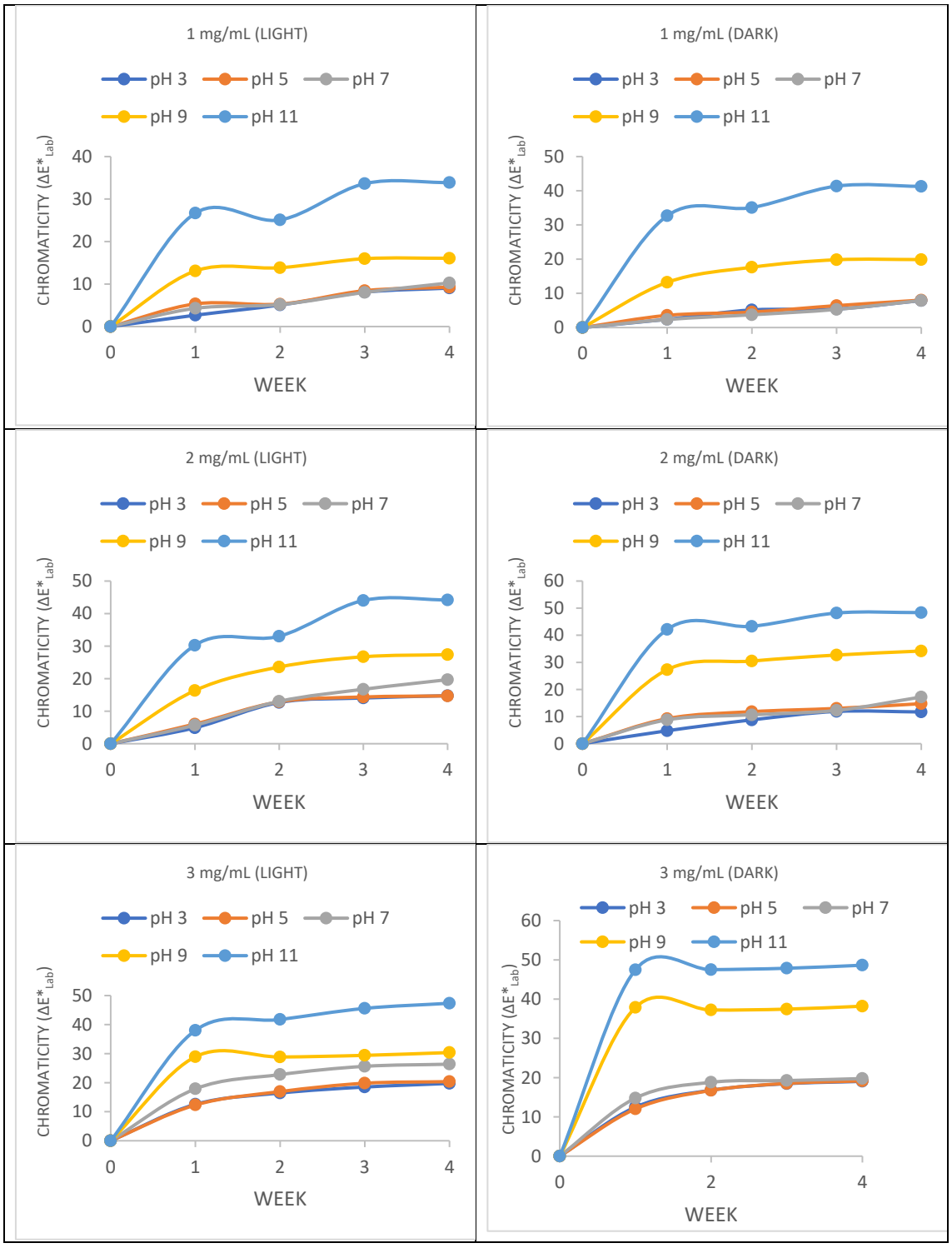

Figure 3. Chromaticity (ΔE^*_{Lab}) values for different concentrations of curcumin extracted from temu lawak (*C. xanthorrhiza*) in light and dark condition

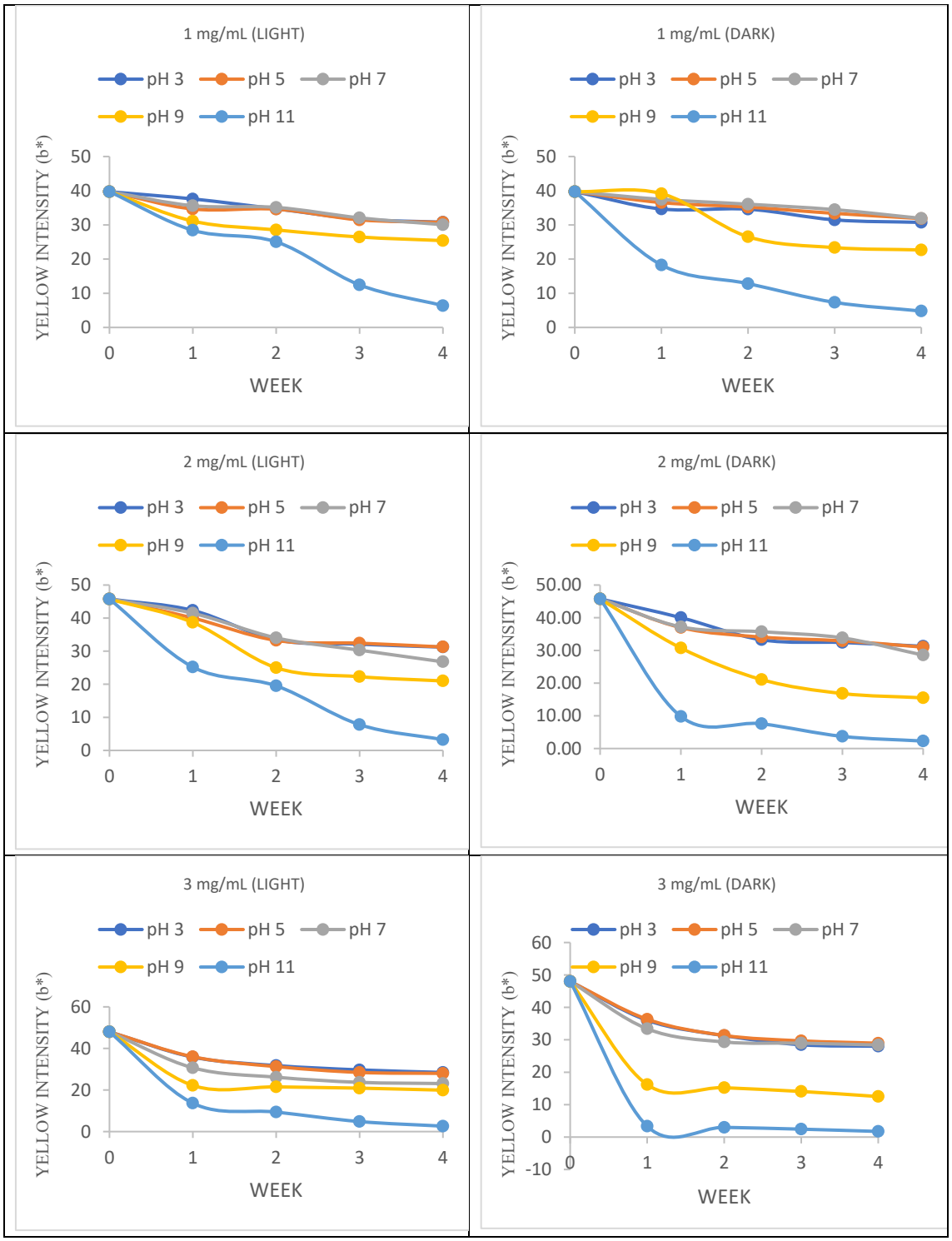

Figure 4. Yellow intensity (b*) values for different concentrations of curcumin extracted from temu lawak (*C. xanthorrhiza*) in light and dark condition for different pH

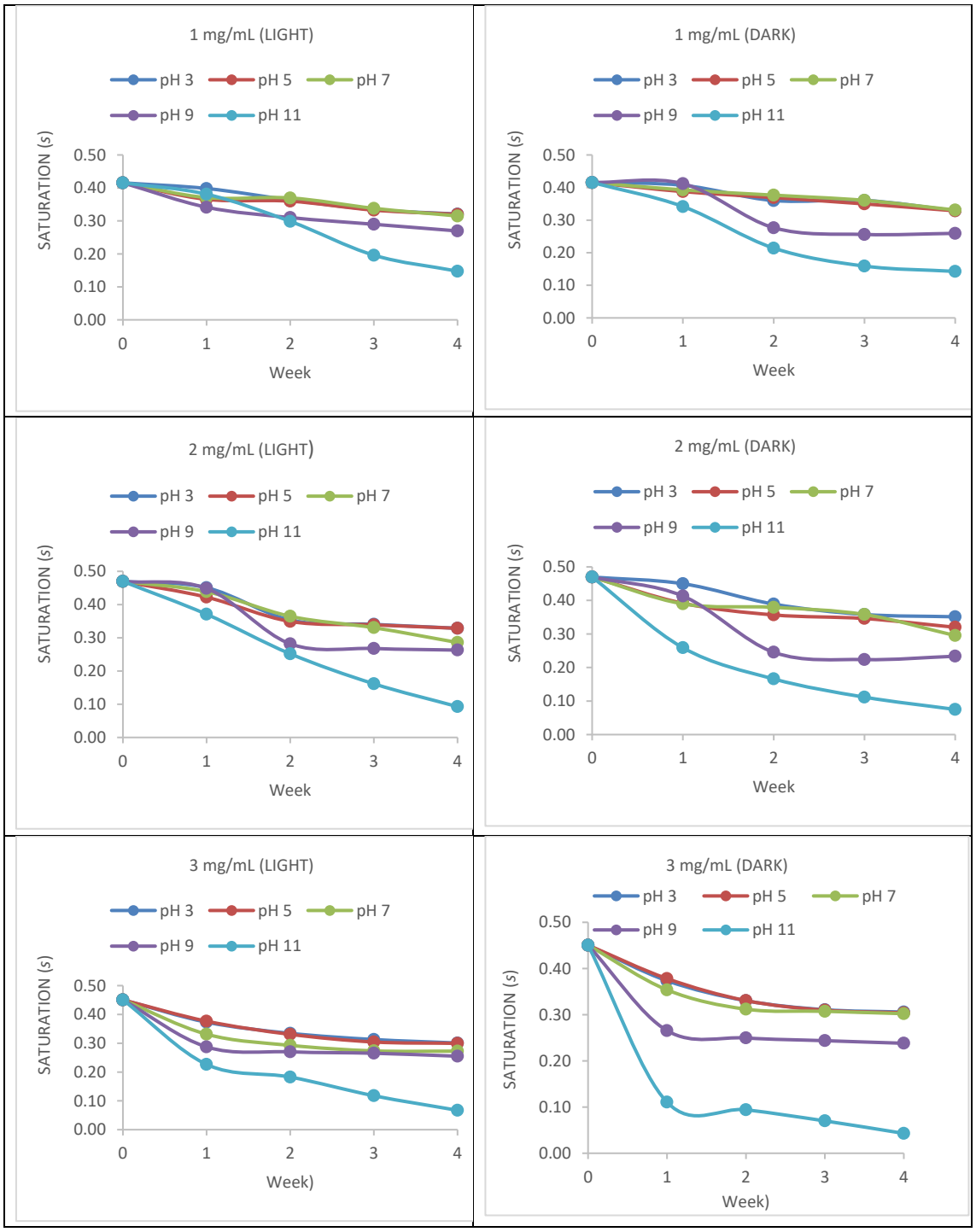

Figure 5. Colour saturation (*s)* values for different concentrations of curcumin extracted from temu lawak (*C. xanthorrhiza*) in light and dark condition for different pH

Effect of UV Irradiation

The ΔE*Lab, s, and b* (%) for different concentrations of curcumin extracted from temulawak and exposed to UV radiation for 8 hours were compared to further investigate the stability of the pigment. The ΔE* results under different types of UV radiation from the beginning until the end of the exposure are shown in Table 2 and Figure 6. The ΔE* corresponds to colour brightness and is generally observed by the colour intensity. In this study, the ΔE* for the curcumin pigment irradiated under UV-A for 8 hours was > 5.0, indicating clearly two different colours, whereas under UV-B irradiation, the ΔE* was < 3.5, indicating a noticeable difference in colour. The s value is a measure

of how colourful an area is visualised by an observer. It is determined as the ratio of ΔE* to L*. As depicted in Table 2 and Figure 6, after 8 hours of exposure, the curcumin pigment exhibited the highest degradation under UV-A as compared to UV-B. At concentrations of 1 mg/mL to 3 mg/mL, its s and yellow intensity degraded from 32.78% to 39.83% and 32.78% to 39.92%, respectively under UV-A, and in contrast, from 8.10% to 3.98% and 6.82% to 5.16%, respectively under UV-B. It was observed that the degradation percentages of s and b* under UV-B decreased as the concentration of curcumin increased, indicating that the curcumin pigment was more stable under UV-B as compared to UV-A. The stability of carotenoids can be impacted by exposure to light (Boon et al. 2010; Rodriguez-Amaya 2001) due to the occurrence of trans-cis photo-isomerisation (Rodriguez-Amaya 2001; Yip et al. 2014). The single O_2 production, that creates the excited state of the molecules of the carotenoid, may also be caused by light absorption. A chemical degradation process might then be involved (Boon et al. 2010; Meléndez-Martínez et al. 2006).

Table 2. Effects of UV radiation for 8 hours on color degradation for different concentrations of curcumin pigment extracted from temu lawak (*C. xanthorrhiza*).

	Degradation percentage (%)					
	UV-A			**UV-B**		
	Curcumin concentration (mg/mL)					
CIE L*a*b*	**1**	**2**	**3**	**1**	**2**	**3**
s	32.78	38.12	39.83	8.10	6.60	3.98
b*	32.78	38.16	39.92	6.82	6.95	5.16
ΔE*	> 5.0	> 5.0	> 5.0	< 3.5	< 3.5	< 3.5
< 3.5 - noticeable colour difference;> 5.0 - indicate clearly two different colours						

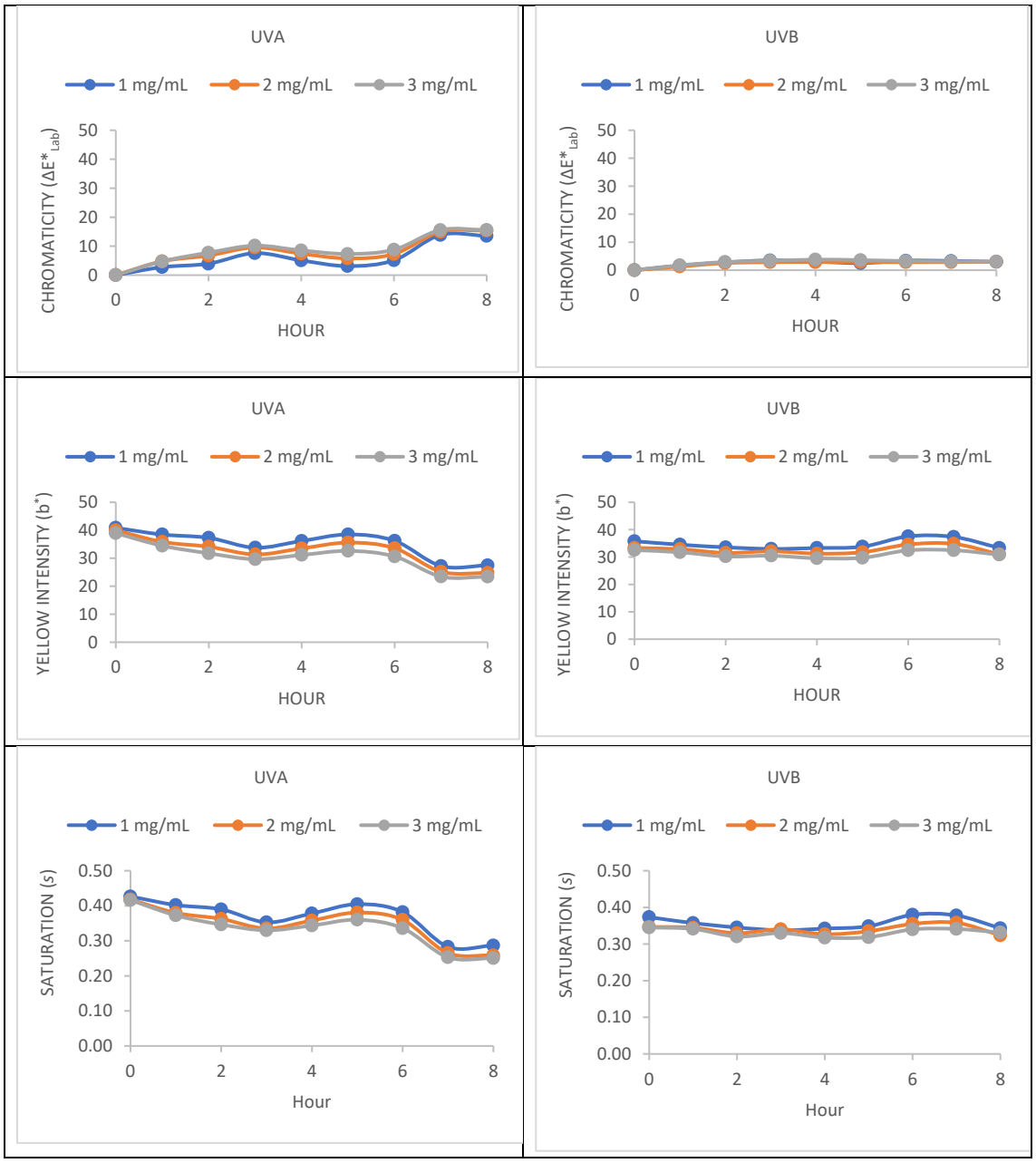

Figure 6. CIE L*a*b* colour difference or chromaticity (ΔE*Lab), yellow intensity (b*), and colour saturation (s) values for different concentrations of curcumin extracted from temu lawak (*C. xanthorrhiza*) and treated with UV radiation for 8 hours

DISCUSSION

pH treatment

Curcumin pigment exhibits stability only in highly acidic conditions, while it becomes unstable at higher pH values. Curcumin concentrations above 1 mg/mL and pH levels exceeding 7 can lead to color degradation and reduced stability, even under dark conditions (Table 1). The results were correlated with those of a previous study by Suresh et al. (2013), which showed that the stability increased in acidic environments and decreased as the pH increased. The decomposition was pH-dependent and proceeded more rapidly in neutral or basic environments. It has been demonstrated that stability increases in acidic pH and decreases in alkaline pH, which is consistent with the findings

of other research, thus indicating that curcumin is more chemically stable in acidic environments (Tonnesen and Karlsen 1985; Wang et al. 1997).

The studies by Kharat et al. (2017) and Anthony et al. (2022) stated that pure curcumin was unstable towards alkaline (pH ≥ 7) and stable in acidic condition (pH < 7). Different pH condition might cause the changes in the molecular structure of the curcumin. Kharat et al. (2017) also proved that curcumin that been stored 30 days in acidic condition only degraded less than 15% while under neutral (pH 7) and alkaline (pH 8) condition, the degradation percentage are about 38 to 47%. The colour shows only minimal changes in acidic but the yellow colour of curcumin was clearly faded after been exposed to alkaline condition. Moreover, Etxabide et al. (2021) mentioned that curcumin also changes to different colour tones such as brown-yellow when exposed to alkaline which in their study at pH > 9. In food packaging industry, curcumin has been used as a colorimetric indicator to notice the existing of alkaline compounds formed during food spoilage as it colours turn from yellow to red when pH changes from acidic to alkaline (Raduly et al. 2021; Luo and Lim 2020; Ma et al. 2020). Thus, it is essential to identify the conditions that regulate the colour changes in curcumin to avoid such differences by taking effective strategies to ensure appropriate stability of curcumin pigment to act as effective pH-indicators (Etxabide et al. 2021).

UV Exposure

Ultraviolet (UV) is one of the solar radiation types which significantly affecting colour stability. UVA which consists longer wavelength is responsible for melanin colour changes while UVB that has shorter wavelength is responsible to damage the protein. The stability of carotenoids can be impacted by light exposure including UV irradiation (Boon et al. 2010; Rodriguez-Amaya 2001), since *trans-cis* photoisomerization can happen (Rodriguez-Amaya 2001; Yip et al. 2014). The single oxygen production that results in the excited state of the carotenoid molecule may also be caused by light absorption. Moreover, each carotenoid consists different number of conjugated double bonds (c.d.b.) which make them a noticeable colour. The longer chromophore of c.d.b., the brighter colour of the pigment. At least 7 c.d.b. are required to have visually be seen while below than 5 c.d.b. are consider as colourless (Mordi et al. 2020; Meléndez-Martínez et al. 2023). For this study, carotenoid from *C. xanthorrhiza* known as curcumin consists at least 7 c.d.b. since it has a yellowish colour in which this organic molecule was able to absorb UV or visible light and led to the rise of colour changes. Previous study confirmed that the delocalization of electrons along the chromophore has contributed to the decrease of molecule energy and lowering the c.d.b. Subsequently, the absorption of UV light can affect the chromaticity degradation of curcumin pigment (Maddah et al. 2020; Mordi et al. 2020). Thus, it can be summarised that, UV radiation is one of factors affecting chromaticity degradation as the observation through colour changes from bright yellow to light yellow.

CONCLUSION

In general, higher concentrations of curcumin, shorter exposure durations, and darker environmental conditions resulted in lower degradation percentages of the pigment. The pigment also exhibited greater stability in highly acidic environments, particularly at pH values below 7. Conversely, at pH levels above 7 and curcumin concentrations exceeding 1 mg/mL, a decrease in pigment stability and an increase in colour degradation were observed, even under dark conditions. Curcumin was found to be more stable under UV-B exposure at higher concentrations compared to UV-A. A significant correlation was identified between the C*, saturation (s), intensity, lightness (L*), and hue angle (H°) values of the pigment and the duration of exposure to both dark and light conditions. These findings support curcumin's potential as a stable Halal-certified natural colorant. It suggests that curcumin pigment extracted from *C. xanthorrhiza* (temulawak) has strong potential as a natural colourant for the halal industry.

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