FE004

Color and Storage Stabilities of Natural Colorant Produced from Sappan Wood

Luxsika Ngamwonglumlert*, Sakamon Devahastin and Naphaporn Chiewchan

Advanced Food Processing Research Laboratory, Department of Food Engineering, Faculty of Engineering, King Mongkut's University of Technology Thonburi, 126 Pracha u-tid Road, Tungkru, Bangkok 10140, Thailand.

Corresponding author: Luxsika Ngamwonglumlert. E-mail: solu_ping@hotmail.com

Abstract

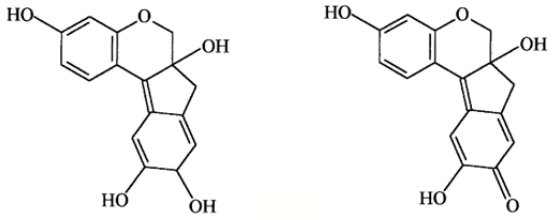
Natural red colorant extracted from sappan wood or heart wood of *Caesalpinia sappan* L. has long been used to prepare foods and beverages. Although this natural dye may have potential as an alternative to synthetic red dye, scientific information, especially on the color and storage stabilities, of this colorant is still limited. In this work, color stability of the wood extract as affected by pH change (at 3, 7 and 9) as well as heat (at 90°C) was investigated. The storage stability of the extract was also assessed at 4°C for 7 days in dark. The results showed that the hue of the wood extract was strongly related to its pH. The extract exhibited yellow color at acidic condition, while exhibiting red color at alkaline condition. The color of the extract was more stable at a lower pH. Change in the color of the wood extract upon heating and storage was related to the changes in lightness and color saturation but was not related to the change in the hue of the wood extract.

Keywords: Acid; Alkaline; Colorant; Heat; pH change; Stability; Storage.

1. Introduction

Sappan wood or heart wood of *Caesalpinia sappan* L. is widely distributed in Thailand, Malaysia, southern China and other regions of Southeast Asia. This wood has normally been used as traditional Chinese medicine (Ohama and Tumpat, 2014; Jin et al., 2015). Since the wood is inexpensive and has no unique flavor (Jin et al., 2015), it also has potential for use as a natural red dye in place of a synthetic dye. The dye extracted from sappan wood has indeed long been added into several foods and beverages (Sinsawasdi, 2012).

Major pigment contained in crude extract of sappan wood is brazilin, which is classified as a neoflavanoid. This pigment, which exhibits colorless or yellow color in nature, is very sensitive to air and light; oxidation of the hydroxyl group of brazilin to a carbonyl group as well as formation of brazilein (de Oliveira et al., 2002; Lioe et al., 2012) as shown in Fig. 1 normally take place. This structural change leads to the change in color of the wood extract from colorless or yellow to red (Eastaugh et al. 2004; Rondao et al. 2013).



Brazilin Brazilein

Figure 1 Brazilin and brazilin structures (Adapted from de Oliveira et al., 2002).

Despite its potential as a natural dye and the fact that it has already been applied in many foods and beverages, scientific information, especially in terms of color and storage stabilities, of this natural dye is still very limited.

In this work, color stability (in terms of L^*, h^*, C^* and ΔE^*) of sappan wood extract against the pH changes (at 3, 5 and 9) and heat (at 90 °C) was evaluated. Storage stability of the extract was also assessed during storage at 4°C in dark for 7 days.

2. Materials and Methods

2.1 Material and reagents

Sappan wood (*Caesalpinia sappan* L.) powder was purchased from a local market in Bang Khae, Bangkok.

Citric acid was purchased from Thai Food and Chemical (Bangkok, Thailand) and sodium hydroxide was purchased from Carlo Erba (Milano, Italy).

2.2 Extract preparation

Water extraction of the wood powder was conducted according to the suggested procedures of Sinsawasdi (2012). About 3 g of the wood powder was added into an Erlenmeyer flask and mixed with 100 mL of distilled water. The mixture was shaken in a shaker (New Brunswick Scientific, G24 Environmental Incubator Shaker, Edison, NJ) at 160 rpm at 30°C for 360 min. The mixture was filtered through Whatman No. 1 filter paper to separate the sediment from the supernatant or the wood extract. The extract was stored at 4°C in dark until the time of the stability test; the storage time was in any case no longer than 1 day.

2.3 Stability evaluation

2.3.1 pH stability

To evaluate the stability of the wood extract against the pH change, pH of the wood extract was adjusted to 3, 7 or 9 by the addition of either 1 N citric acid or 1 N sodium hydroxide solution. Adjusting the pH of the wood extract to 3, 7 and 9 represents the use of the extract as a dye in acidic, neutral and alkaline foods, respectively. The color (in terms of L*, h* and C*) of each pH-adjusted extract was measured.

2.3.2 Thermal stability

The pH of the wood extract was first adjusted following the method in Section 2.3.1. About 25 mL of each pH-adjusted extract was then filled in a series of test tubes, closed with screw caps and heated in a water bath at 90°C. Tubes containing the wood extract at pH 3, 7 or 9 were removed from the bath after heating for 60 min and rapidly cooled in an ice-water mixture prior to the color measurement (in terms of L^*, h^*, C^* and ΔE^*). The pH-adjusted extract without heating was used as a control sample.

2.3.3 Storage stability

Each pH-adjusted extract was kept at 4°C in dark in a refrigerator for 7 days. After 7 days, color (in terms of L^*, h^*, C^* and ΔE^*) of the extract was measured to evaluate its cold storage stability. The pH-adjusted extract prior to the storage was used as a control sample.

2.4 Color measurement

Color of the extract was measured in the CIELAB color space via the use of a spectrophotometer (HunterLab, ColorQuest XE, Reston, VA). The measurement was performed in a transmittance mode using a simulated D65 illuminant and 10° observer angle. The hue angle (h^*), chroma (C^*) and total color difference (ΔE^*) were calculated using Eqs. (1) to (3).

$$h^* = \tan^{-1}\left(\frac{b_i^*}{a_i^*}\right) \quad (1)$$

$$C^* = \sqrt{\left(a_i^*\right)^2 + \left(b_i^*\right)^2} \quad (2)$$

$$\Delta E^* = \sqrt{(L_i^* - L_0^*)^2 + (a_i^* - a_0^*)^2 + (b_i^* - b_0^*)^2} \quad (3)$$

where L_i^*, a_i^* and b_i^* are the lightness, redness/greenness and yellowness/blueness of any sample; L_0^*, a_0^* and b_0^* are the lightness, redness/greenness and yellowness / blueness of the control sample.

2.5 Statistical analysis

The experimental data were subject to the analysis of variance (ANOVA) using a statistical program Minitab (version 16) and are presented as mean values with standard deviations. Differences between mean values were established using the least significant difference (LSD) tests at a confidence level of 95%. All experiments were performed at least in triplicate.

3. Results and Discussion

3.1 Color stability against pH change

The effect of pH change on the color (in terms of L^*, h^* and C^*) of the wood extract is shown in Fig. 2. Colors of the extracts at pH 3, 7 and 9 were yellow, orange and red, respectively. Besides air and light, pH is also one important factor affecting the change in the color of the wood extract. At acidic condition, the wood extract is predominated by yellow-colored brazilin. Increase in the pH induces the deprotonation of brazilin, resulting in structural transformation to brazilein (Rondao et al., 2013) and hence the color change to orange and red at pH 5 and 7, respectively. Similar results were also reported by Rina et al. (2017) who found that the wood extract exhibited

red color when added into carbonated drink (pH 9) and yellow color when added into orange juice (pH 3).

The above results confirm that the wood extract is highly sensitive to pH change. Thus, when the wood extract is used as a food dye, pH of food must be first checked if the the expected color is to be obtained. However, it is interesting to note that the direction of the color change of the wood extract is opposite that of anthocyanins, which changes from red to yellow when the pH increases (Ngamwonglumlert et al., 2017). Mixing of the wood extract with anthocyanin extract may yield a new dye, which may be stable over a wider range of pH.

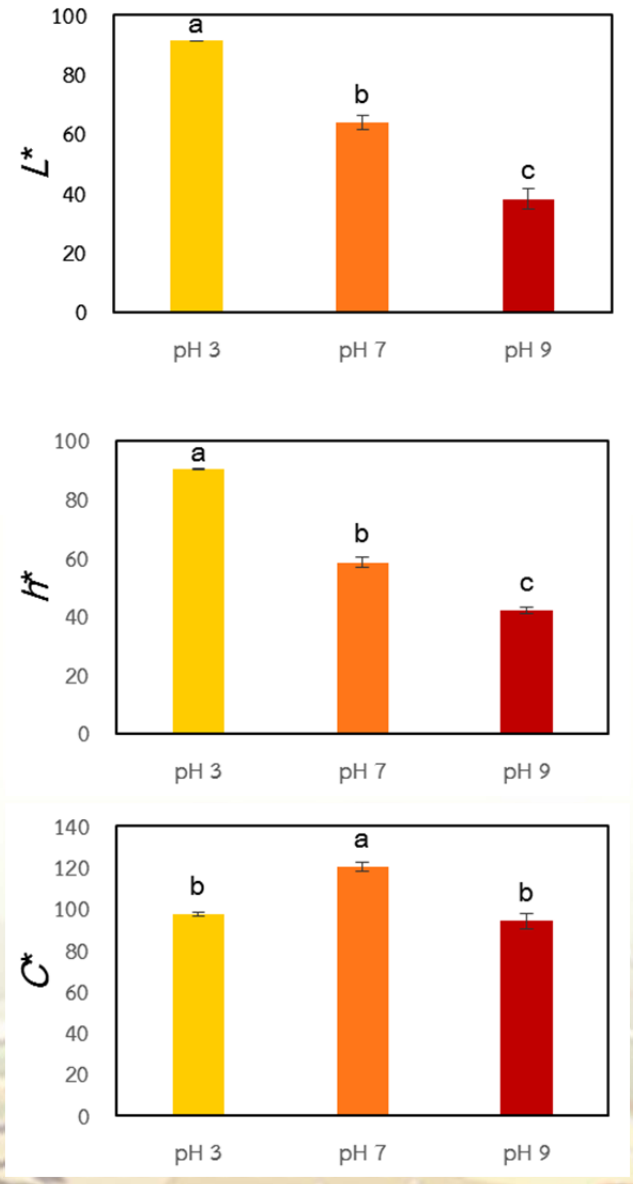
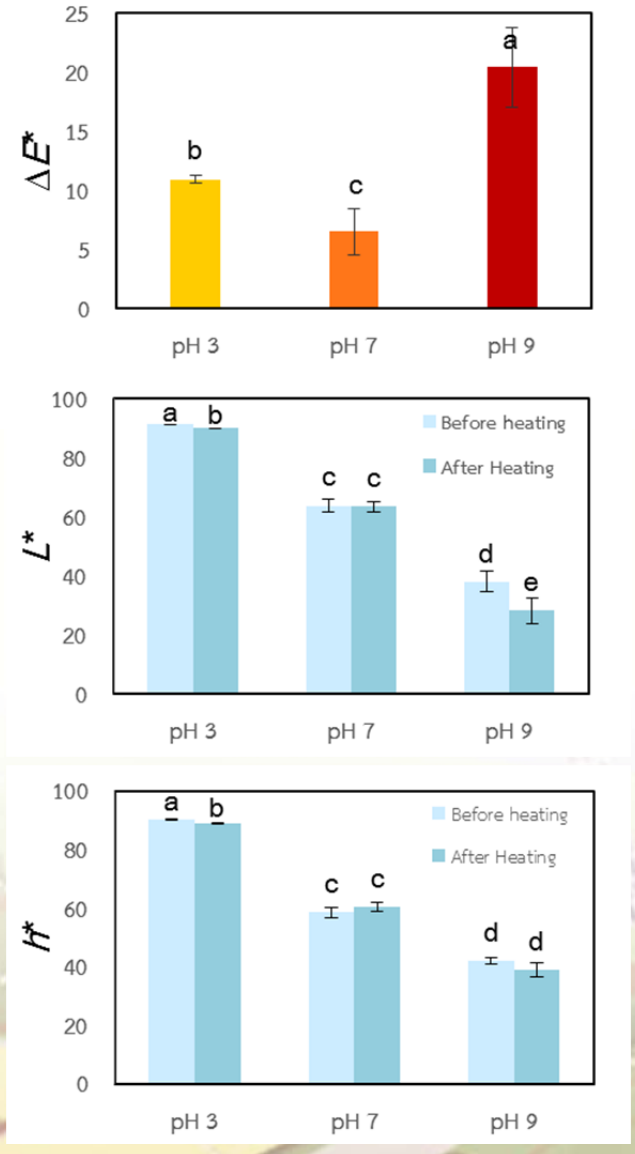


Figure 2 Color (in terms of L*, h* and C*) of the wood extracts at different pH. Different letters over the bars indicate that values are significantly different ($p < 0.05$).

3.2 Color stability against heat at different pH

The effect of heat on the color (in terms of *L**, *h**, *C** and ΔE^*) of the wood extract is shown in Fig. 3. The results showed that the wood extract at pH 9 had lower stability against heat than at pH 3 and 7 as can be seen through the higher in ΔE^* values. Changes in *L** and *C** values, which represent the changes in lightness and color saturation of the wood extract, were clearly observed after heating, while *h** value only slightly changed. Changes in *L** value upon heating might be due to the oxidation of polyphenols contained in sappan wood, which resulted in the formation of dark compounds (Jin et al., 2015; Liu et al., 2009) and hence the decrease in *L** value of the extracts.



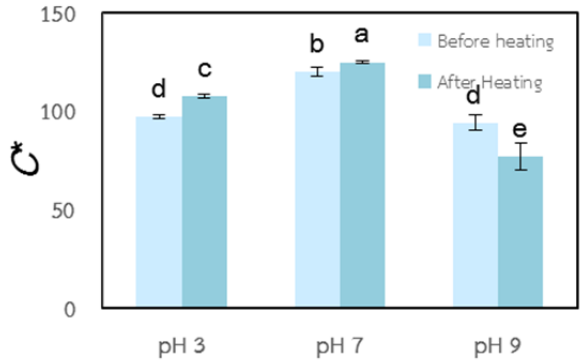
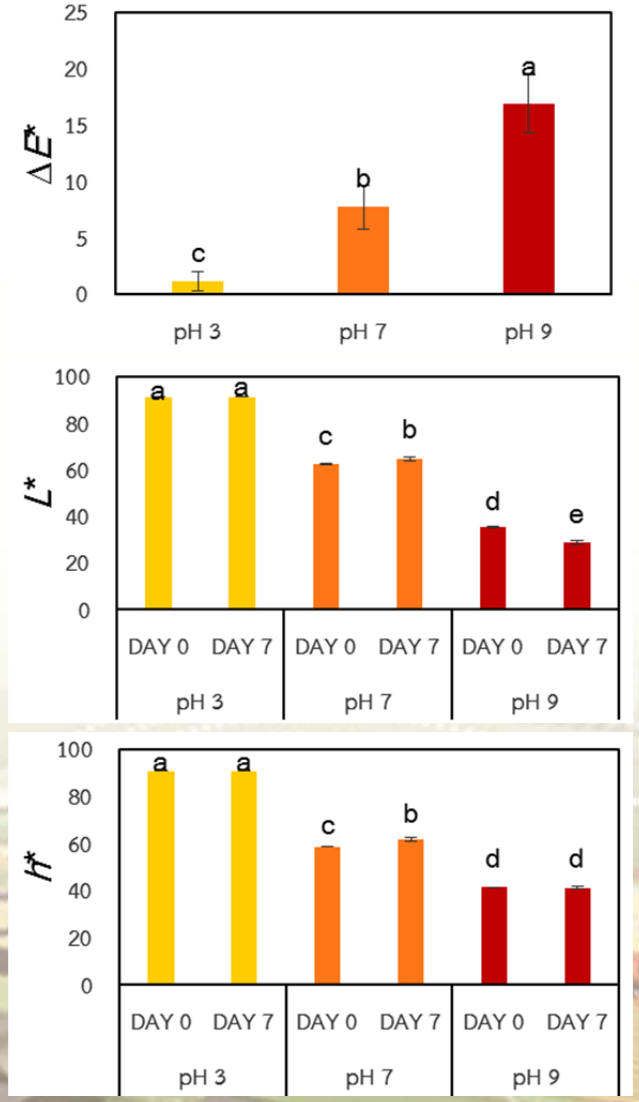


Figure 3 Color stability of the wood extracts against heat (90°C) at different pH. Different letters over the bars indicate that values are significantly different ($p < 0.05$).

3.3 Storage stability.

Change in the color of the extracts at different pH upon storage for 7 days at 4°C in dark is shown in Fig. 4. The extract at pH 9 possessed lower stability upon storage than at pH 7 and 3, respectively.



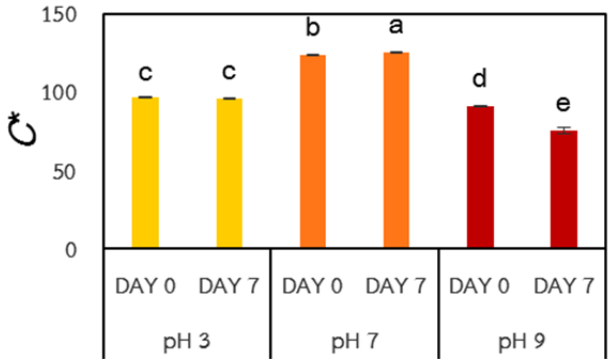


Figure 4 Color of the wood extracts at different pH both before and after storage at 4°C in dark. Different letters over the bars indicate that values are significantly different ($p < 0.05$).

Change in the color of all extracts did not correspond to the change in h^* values as the extracts still exhibited yellow, orange and red color at pH 3, 7 and 9. The color changes of the extracts at pH 7 and 9 upon storage were the result of the changes in L^* and C^* values, which may be caused by the degradation of red pigment or brazilein.

4. Conclusions

The pH, thermal and storage stabilities of sappan wood extracts were investigated. Change in pH induced change in the hue of the extracts. The extracts exhibited yellow, orange and red color when the pH was adjusted to 3, 7 and 9, respectively. Heat (90°C) did not affect the change in the hue of the wood extract but affected the changes in lightness and color saturation. Hue of the wood extract also did not change upon storage for 7 days at 4°C in dark, while lightness and color saturation changed. The extract at pH 9 possessed lowest thermal and storage stabilities as it had the highest ΔE^* values. Further investigation on the degradation and transformation of brazilin and brazilein during heating and storage is needed to clearly understand and explain the results.

5. Acknowledgements

The authors express their sincere appreciation to the Thailand Research Fund (TRF, Grant no. RTA 5880009) for supporting the study financially. Author Ngamwonglumlert thanks the Royal Golden Jubilee (RGJ) Ph.D. Program of the TRF and the Petchra Pra Jom Klao Ph. D. Research Scholarship of King Mongkut's University of Technology Thonburi for supporting her doctoral study.

6. References

de Oliveira, L.F.C., Edwards, H.G.M., Velozo, E.S., Nesbitt, M. 2002. Vibrational spectroscopic study of brazilin and brazilein, the main constituents of brazilwood from Brazil. Vibrational Spectroscopy 28, 243–249.

Eastaugh, N., Walsh, V., Chaplin, T., Siddall, R. 2004. The Pigment Compendium: A Dictionary of Historical Pigments. Burlington, MA: Elsevier.

Jin, S.K., Ha, S.R., Choi, J.S. 2015. Effect of Caesalpinia sappan L. extract on physico-chemical properties of emulsion-type pork sausage during cold storage. Meat Science 110, 245–252.

Lioe, H.N., Adawiyah, D.R., Anggraeni, R. 2012. Isolation and characterization of the major natural dyestuff component of Brazilwood (Caesalpinia sappan L.). International Food Research Journal 19, 537–542.

Liu, D.C., Tsau, R.T., Lin, Y.C., Jan, S.S., Tan, F.J. 2009. Effect of various levels of rosemary or Chinese mahogany on the quality of fresh chicken sausage during refrigerated storage. Food Chemistry 117, 106–113.

Ngamwonglumlert, L., Devahastin, S., Chiewchan, N. 2017. Natural colorants: Pigment stability and extraction yield enhancement via utilization of appropriate pretreatment and extraction methods. Critical Reviews in Food Science and Nutrition 57, 3243–3259.

Ohama, P., Tumpat, N. 2014. Textile dyeing with natural dye from sappan tree (Caesalpinia sappan Linn.) extract. International Journal of Chemical, Molecular, Nuclear, Materials and Metallurgical Engineering 8, 432–434.

Rina, O., Ibrahim, S., Dharma, A., Utami, C., Widodo, Y.R. 2017. Stabilities natural colorant of sappan wood (Caesalpinia sappan. L) for food and beverages in various pH, temperature, and matrices of food. International Journal of ChemTech Research 10, 98–103.

Rondao, R.J., de Melo, J.S.S., Pina, J., Melo, M.J., Vitorino, T., Parola, A.J.D. 2013. Brazilwood reds: The (photo)chemistry of brazilin and brazilein. Journal of Physical Chemistry A 117, 10650–10660.

Sinsawasdi, V.K. 2012. Sappanwood water extract: Evaluation of color properties, functional properties, and toxicity. PhD dissertation. Gainesville, FL: Department of Food Science and Human Nutrition, University of Florida.