Available online on 15.10.2020 at http://ajprd.com

Asian Journal of Pharmaceutical Research and Development

Open Access to Pharmaceutical and Medical Research

Open Access

Review Article

The Degradation Rates of Natural Dyes from Natural Resources: A Review

Yesi Armalina, Zikra Azizah, Ridho Asra*

School of Pharmaceutical Science (STIFARM) Padang, Indonesia 25147

A B S T R A C T

Objectives: Natural plants can produce natural dyes. These dyes are widely used in the pharmaceutical and food fields, such as betacyanin, anthocyanin, carotenoids, flavonoids and others. Several external factors could cause natural dyes to show instability or degradation, some of the external factors are temperature, pH, storage time and so forth. Research on various kinds of natural dyes shows the degradation of these natural dyes. Degradation can cause damage to these plants dyes, therefore steps are needed to prevent the degradation of these dyes. The measurement of degradation was carried out to prevent the deterioration of natural dyes from natural materials that will be used or consumed. The purpose of this review is to determine the rate of degradation of natural dyes sourced from nature in accordance with the results of the studies that have been conducted.

Data Sources Study Selection: Data sources of this review article taken from web Google Scholar, Science Direct, National Center for Biotechnology Information (NCBI), Scopus, and Pubmed.

Conclusion: Natural dyes can be used for manufacture of food or drinks, natural dyes from plants instability due to degradation. Degradation occur because temperature, pH, storage time and sunlight.

Keywords: Activation energy, Degradation, Dyes, Half-life, Natural substances, pH, Reaction order, Storage time, Temperature.

A R T I C L E I N F O: Received 25 June 2020; Review Completed 13 Sept. 2020; Accepted 02 Oct. 2020; Available online 15 Oct. 2020

Cite this article as:

Yesi A, Zikra A, Ridho A, The Degradation Rates of Natural Dyes from Natural Resources: A Review, Asian Journal of Pharmaceutical Research and Development. 2020; 8(5):75-81. **DOI:** http://dx.doi.org/10.22270/ajprd.v8i5.827

***Address for Correspondence:**

Ridho Asra, School of Pharmaceutical Science (STIFARM) Padang, Indonesia 25147

INTRODUCTION

Natural dyes are available in nature in various colors[1], and are the main characteristic of any food ingredient because they can increase attraction[2]. Dyes can be produced from plants, animals and microorganisms. In plants, dyes can be produced directly or indirectly because they have pigments. Pigment is a component that can absorb UV light at a certain wavelength range[3,4]. The physical properties of natural dyes can be well understood based on the wavelength of the dye based on the appearance of the corresponding color[5].

Natural dyes are an important alternative because they are considered harmless to health[6]. Natural dyes also have an important role in human health because they have biological components and pharmacological properties[7] such as carotenoids acting as photo-protectors, antioxidants, precursors and plant hormones[8], betalanine has a tendency to antioxidants[9], betacyanin and betaxanthin are used as natural dyes in food[10]. Natural dyes are widely used in industries such as anthocyanins[11], but natural dyes have a disadvantage, namely their instability to processing[12].

Natural dyes can show a chemical composition that affects stability, solubility and are also very sensitive to pH, temperature and sunlight[1]. Effect of temperature is a factor influencing the degradation kinetics[13], the kinetics of color loss and pigment degradation can be calculated for each color[14]. The color degradation studied is related to the spectral properties and visual color[15].

Visual color is monitored from the CIEL, a*, b* parameters. The absorbance at maximum wavelength /total

col was used for quantitative color degradation[16]. Chroma (c) and hue (h) were estimated by values a* and b *[17]. Color parameters such as color density, color intensity and brownish index follow the total substance content[18]. Dye parameters take into account the chemical or degradation kinetics approach by calculating the kinetics parameters of the reaction order, reaction rate constant (k), half-life (t½), activation energy required for the determination of degradation[19,20].

Color analysis for visual color parameters using Hunter Lab coordination was used to calculate L, a *, b * values where L * L* (brightness, 0 from black to 100 for white), value a* (red-green) and value b* (yellow-blue)[21,22]. Differences in kinetics models are also evaluated to explain the changes that occur. The kinetics models consist of zero-order reaction, first-order reaction, second-order reaction, and fractional conversion kinetic model[23]. The value of the degradation constant (k) is obtained from the plot line logs percent of substance retention with time[24]. Reaction rate can be determined from the biggest value of R^2. The larger the value of R^2 the easier the observation would be done following the reaction order, the value of Ea or activation energy shows that the value of k can be sensitive to changes in temperature and the value of k obtained from equation can be used to calculate the half-life, while the value of activation energy (Ea) is obtained from connecting the values of In k and 1 / T[25].

Kinetic models are often used for objective, rapid and economical food safety assessments. Knowledge of degradation kinetics including reaction order, rate constant, activation energy is very important to predict food quality loss during storage as well as heat treatment[26]. A mathematical model has been developed using kinetic parameters obtained from isothermal experiments to predict color loss in isothermal or non-isothermal methods[27].

Recent studies suggest hyperbaric storage at room temperature (HS-RT) can be an attractive method for food storage or preservation. The effect of HS-RT was carried out to evaluate the dye degradation in strawberry juice[28]. Studies conducted using HSST on beet stems were considered to be the most suitable for maintaining the stability of betalain[29]. A study conducted by[30] with the addition of acid to the sample resulted in color degradation due to reduced concentration and a change in color. In another study it was said that tartaric acid and citric acid decreased monomeric anthocyanin content, brightness, increased polymer color index, browning degradation, chroma and red and blue color parameters, and hue angle[31]. From this explanation, this review article will explain the degradation of dyes from natural materials that have been studied several years previously.

METHODS

Articles review were carried out in journals related to the degradation of natural dyes of natural origin. The journal was obtained by searching on several quality scientific web sites based on scientific publication guidelines, internationally accredited, in the international, and national categories. The keywords used to search for research journals were "degradation", "degradation kinetics", "natural dyes", "degradation of natural dyes", and "degradation of natural materials". The scientific web used were Google Scholar, Science Direct, National Center for Biotechnology Information (NCBI), Scopus, and Pubmed. The search for research journals for review articles had criteria as follows: that the research journals should not have been published for more than 10 years, that the research journal conducted research on the degradation of natural dyes in natural materials, the research journals have knowledge related to degradation, natural dyes, natural ingredients and so forth. This review article was conducted in research journals by looking at the analysis of dyes that occur in natural ingredients or food / beverage products on the influence of temperature, pH, sunlight and storage time.

RESULTS AND DISCUSSION

A. Degradation on natural dyes

Dragon fruit extract studied[32] showed absorbance peaks of 230 nm and 537 nm on UV-Vis spectrophotometry. The absorption peak showed the presence of a dye, namely betacyanin. Changes in the intensity of betacyanin were caused by storage conditions, light, temperature, pH and additive properties for 3 weeks at 537 nm. The degradation test of red dragon fruit dye at a temperature of 40^0C showed that there was no color change with a degradation value of 11.54%, while the test at 100^0C showed that the color change in betanine was disappearing with the degradation value increasing by 84.23%[33]. Dragon fruit peel dye degradation tests[34,35] in pH treatment showed a decrease in absorbance where the absorbance value began to decrease at high pH and the macerate color changed from red to a more faded color. At high pH there is damage to betacyanin because the compounds in the macerate are rapidly hydrolyzed. Testing of red dragon fruit peel dye in sunlight showed a change in the color of the sample, changing from a red color to a more faded color. In addition, there was a decrease in the absorbance of the sample so that the color of the sample became unstable[36].

The dye degradation test was related to the temperature of different fruit, namely the blackberry fruit which degraded at at 70-75^{0}C and 80-90^{0}C. The degradation did not decrease too much at 70^0C, but after 90 minutes there was a decrease as much as 20%. At high temperatures of 80-90^{0}C there was a degradation of 40% during 90 minutes of heating. The order of anthocyanin reactions from blackberries followed order 1 because the k values obtained were from 0.0155 to 0.0051 min^{-1}, the activation energy (Ea) values of heating at ocmic and conventional were 67 kJ/mol and 56 kJ/mol[37].

The dye degradation test on roselle flowers[38] showed that the extract that was added with HCl decreased by 22% at a temperature of 70-80^{0}C and experienced a slight decrease in storage time. The extract that was added with formic acid decreased by 35% at a temperature of 70-80^{0}C. and an increase in storage time by 11%. The extract added with citric acid was almost the same as formic acid, where at a temperature of 70-80^{0}C the decrease in storage time was 44% and was followed by a 60% decrease at 85^0C. The extract added with acetic acid increased by 23% at 80 - 85^0C. In heat degradation testing of the roselle anthocyanin in a buffer solution with a pH of 1-7 following the first-order reaction. The half-life values (t½) of anthocyanin

degradation were 115.2, 385.1 and 182.4 minutes at pH 1 with a temperature of 70^0C, 80^0C and 90^0C, while at pH 7 the half-life values (t½) of anthocyanin degradation were 13.5, 19.4 and 24.8 minutes at temperatures of 70^0C, 80^0C and 90^0C. Activation energy (Ea) in highly acidic materials had a range of 55.8-95.7 kJ/mol. However, the range for the non-acidic material was 31.4-94.7 kJ/mol with the lowest activation energy (Ea) at pH 4 at 55.8 kJ/mol

The purple sweet potato peel extract was tested for the combined degradation of temperature and pH[40]. The temperature treatment test did not show any degradation or change of the extract. In the pH treatment test, namely pH 1 there was no degradation, while pH 3 and 5 there was degradation with values of 49.63% and 52.52%. Tests were also carried out on the heat degradation of purple sweet potato in aqueous solution with a difference in pH[41], it was found that the half-life value (t½) at 80^0C increased significantly from 41.8 hours at pH 2 to 96.3 hours at pH 3 and decreased rapidly to 8.9 hours at pH 6. The values of activation energy (Ea) during heating at pH 2-6 are 99.95, 111.57, 84.87, 66.56 and 89.39 kJ/mol. Anthocyanin degradation during storage following the purple sweet potato first order reaction indicated by the value of R^2.The half-lives (t½) of anthocyanins from purple sweet potato paste were 119.51, 60.80, 32.85 days at 30^0C, 40^0C, and 50^0C. Anthocyanin degradation during storage was also indicated by color changes. This change became better with increasing storage temperature. The activation energy (Ea) for anthocyanin degradation at each temperature increase of 10^0C is 52.60kj/mol. Meanwhile, the increase in activation energy (Ea) for the change in H* and C* for each temperature change of 10^0C is 55. 83 kj/mol and 49.99 kj/ mol[42].

The dye on the peels of eggplant, grapes, plum, strawberry and billberry skins was also tested for degradation by storing it for 17 weeks at a pH of 4.5 and a temperature of 20°C. on eggplant peel by 61%, billberry fruit by 64% for 17 weeks of storage. However, during storage on strawberries, anthocyanins lost their concentration at week 4 by 81% and week 17 by 99%. The highest L * values were obtained between 26.59 and 29.56 for strawberries and grapes. In the storage of all of the above fruits, the L * value is only slightly reduced by about 1-4%. The value of R^2 ranged from 0.9470 to 0.9855 indicates a good correlation between anthocyanin concentration and storage time. The stability of half-life (t½) and the highest D values at 12 and 17 weeks of storage were shown by eggplant peel, grape, and plum peel. Meanwhile, the low stability was indicated by the strawberry fruit which had a half-life (t½) and a D value of 2.8 with 4 weeks of storage[43].

Purple corn with color parameters L*, a*, b* on PCW, EA, F1 and F2 dissolved in citrate buffer solution with pH variations at 22^0C was tested for a month to evaluate color changes with a storage period of more than 12 weeks. The test results at pH 5 and 6 showed a fuzzy color. Whereas at pH 3.5-6 at time 0, the color has increased in yellow hue. All treatments with increasing pH showed an increase in L, E * ab, but a*, b*, hue and chroma values decreased. Chroma changes occurred at pH 2.0, 2.5, and 3.0 during 12 weeks of storage. A slight color difference was observed at pH 2 (ΔE from 0.2 to 3.6). After 12 weeks, pH 6 caused a change in color (ΔE from 17.7 to 47.5) and a decrease in the estimated half-life (t½) of total anthocyanins (range 1.8 to 3 weeks), compared with pH 2 (44.6 to 60.7 weeks)[44].

Broccoli frozen before cold storage had an L=-22.0±1.91, a*=-17.41 = 0.75, b*=15.97±1.13, and h*=137.95= 3.91. In the isothermal storage conditions, the temperature of -7^0C broccoli became brighter, the L value increased 53%, the a* (a*/ao*) and h values decreased. Color degradation parameter a*/ao* and b explained the arhernius order kinetics models 0 and 1 similarities. The value of a and b, including models that correspond to the stage regression analysis, coefficient value of R^2 determination is high (0.85 in two situations). Under nonisothermal storage conditions (57 days), the color parameters of a*/ao* and h broccoli decreased by 30 and 5%. Alteration was very low compared with storage at -7^0C for the same time (a*/ao* degraded 47% and h 13%) but higher at -25^0C (a*/ao* degraded 17% and h* 3%). In the conditions under nonisothermal kinetics models of order 0 and 1 meeting the conditions for changes in a*/ao* and h, the coefficient of determination is 0.40 and 0.44 for a*/ao* and h. The color kinetics of broccoli alteration under nonisothermal storage has low activation energy (Ea) values[45].

"Merlot" and "Ruby" grapes were stored at 5^0C, 25^0C and 30^0C for 360 days. Merlot juice had a higher anthocyanin content ($p < 0.005$) than ruby juice. During storage, the individual total anthocyanins followed the 1st order reaction model. At the end of storage, higher anthocyanins (95-99.9%) were observed at 25^0C and 35^0C compared tostorage 5^0C "merlot"(50-60%) and "Ruby" (74-81%). Anthocyanin compounds that were most sensitive to temperature are C3G (Ea = 66.5 kj/mol) and D3G (Ea = 63.3 kj/mol). The half-life (t½) at 5^0C for C3G was 73 days and D3G was 69 days[46].

Anthocyanin stability from cranberrybush fruit, in water and ethanol extracts, which were stored in the dark for 7 days at different temperatures (2^0C, 37^0C and 75^0C) at pH 3 and 7 had also been tested. The test results showed that the lowest anthocyanin stability was found in water extracts stored at 75^0C with a pH of 7, with a half-life and constant velocity values of 1.98 hours and 0.3488 / h. In addition, the test also showed a good correlation between anthocyanin content and storage time with R^2 = 0.9298-0.9971 and following the 1st order reaction model with all test conditions[47].

Extremely acidic papaya peel extract (pH = 4) showed a very low content of betalain, chroma (C*ab = 60 against 70) and a yellow color component (h*). The storage temperature shows a tendency towards a red hue (hue from 10 to 80) and a change in brightness L* (90 to 75) as the temperature increases. At lower acid levels, pH 5 and 6 are superior to the angle of color stability[48].

Color analysis on fresh moringa leaves, respectively, had L *,a*,b* values, namely 40.78, -4.20 and 6.33. The brightness values of L* are 2.46, 3.10 and 4.26 at 40^0C, 50^0C and 60^0C decreasing as the temperature increases and the color became darker. Dry leaves at all temperatures showed an increase in the a* values were 0.39, 0.51 and 1.06 at 40^0C, 50^0C and 60^0C. The b* values at each

increase in drying temperature were 5.13, 6.54 and 5.63 at 40^0C , 50^0C and 60^0C an increase in the b* value in temperature indicated the leaves were turning yellow[49].

In addition to testing on the above plants, research was also carried out on color changes in dried longan meat. The longan meat was dried with a drying air temperature of 60-130^0C. The kinetic parameters for color change were determined using the CIE system. Zero-order reaction kinetic model was most appropriate for describing a* and effective to present changes in ΔE. In addition, the drying temperature and drying time also affected the color change during drying. During drying the L* value decreased and the temperature rose. The a*, b* and ΔE values increased in the first period, then decreased with time and drying temperature[50].

The acerola pulp studied showed L* = 44.07 ± 0.41, a* = 7.84 ± 0.22, and b* = 20.05 ± 0.51, where the character of acerola pulp was orange / red. The standard deviation was more than 2.5 and 6% for parameters L*, a*, b*. Color parameters were in accordance with the model of the first order reaction. The coefficient of determination ($R^{2)}$ was shown with a higher value, the value of 0.89 for all treatments. L*, a*, b* values decreased over time for all treatments[51].

Meanwhile, the color degradation of bael fruit with an average value of L and b at 0 time were 56.98, 32.07, and 59.34, respectively. The values for L, a*, and b* for infinite time are 3.55, 4.02 and 4.55. The value of L decreased from 53.12 to 45.15, 43.11, 37.96 for temperatures, respectively, were 30^0C, 44^0C, 60^0C and 75^0C. The a* value decreased from 30.96 to 26.13, 27.17, 24.14 and 20.99 at temperatures of 30^0C, 45^0C, 60^0C and 75^0C. The value of b* also decreased from 56.25 to 49.16, 48.96, 44.18 and 42.80 at temperatures of 30^0C, 45^0C, 60^0C and 75^0C The decrease in L*, a*, b* values occurred because of an increase in temperature or heat treatment. Value degradation of the first order reaction were 0.07, 0.11, 0.018 and 0.023 to a temperature of 30^0C, 45^0C, 60^0C and 75^0C. The activation energy (Ea) at Bael fruit pulp was 23.83 kJ/mol with a value of $R^2 = 0.9897$[52].

The degradation of green color on rocket leaves followed the first-order reaction. The R^2 value was more than 0.98 and the standard error was less than 0.009. Value constant visual color temperature followed the arhernius equation ($R^{2>}$0.98). The activation energy (Ea) and the frequency factor (ko) for color degradation of the leaves were 51.21 kj / mol and $3.22x10^7$ h^{-1} [53]. The heat degradation of green vegetables followed the first order reaction. The activation energy (Ea) for green color in leaves was 88.78 kj / mol[54]. Carotenoid kinetics and color degradation and furosin formation were investigated in apricots during heating at 50^0C, 60^0C and 70^0C. Carotenoid degradation and color were expressed as total color difference (TCD) followed by the zero-order and the first-order reaction. Activation energy (Ea) for carotene degradation ranged from 73.7 kj/ mol for 13-sis-B-carotene to 120.7 kj/mol for lutein, about 91 kj/mol and 97 kj / mol for all tras-b-carotene and TCAR. While the activation energy (Ea) for TCD change was 67 kj /mol[55].

The dye degradation test on mullberry fruit was carried out at 60^0C, 70^0C and 80^0C. The degradation of anthocyanins decreased by 56.02%; 83.74%; 91.67% at 60^0C, 70^0C, 80^0C for 600 minutes. Heat degradation of mullberries follows the first-order reaction, related to temperature. The temperature will increase the value of k1. Half-life value (t½)from anthocyanin degradation at temperatures of 60^0C, 70^0C, and 80^0C is 8.3; 4,4; 3.2 hours with an activation energy value (Ea) of 46.32 kJ/mol[56].

The dye degradation test on chesnuts was carried out at a temperature of 20-70^0C. Hail, the dye degradation test showed that the color change in chesnuts was darker than the original color. This happened because the value of L* decreased around 26 at 60^0C, 65^0C and 70^0C for 30, 10 and 5 minutes. For comparison, the zero-order reaction ($R^{2=}$0906), the 1st order reaction ($R^{2=}$0946) were in accordance with the data of dye at 5 different temperatures. The five temperature were 50^0C, 55^0C, 60^0C, 65^0C, 70^0C. The activation energy value (Ea) of chestnut color was greater than that of other fruits and nuts, namely L* = 287.19 kJ mol-1 and b* = 347.48 kJ mol-1 during heating treatment[57].

Dye degradation tests on cactus fruit extract [58] showed a decrease in betacyanin at various pHs. The pH tested started from pH 3, 4, 5, 6, 7 and 8. The decrease in betacyanin at that pH was 16.95%; 15.82%; 14,255%; 15.71%; 19.27% and 35.11%. At pH that was too low or high, there was instability in the extract group which results in a color change in the cactus fruit extract.

The degradation test of brazilien dye on secang wood showed that there was pigment degradation due to sunlight which was marked by a decrease in absorbance. The absorbance was visible by changing the pigment to translucency and the red color became invisible. This was because sunlight contained ultraviolet with large energy causing photochemical reactions so that the dye became unstable[59].

The degradation test of the *banggai* sweet potato dye in clear and dark vials showed degradation which was shown by the change in the color of the extract, the longer the irradiation time the more faded the color of the extract was. In addition, there was also an hourly increase in degradation, where the longer the exposure to sunlight, the greater the level of degradation would be. The resulting levels of sun exposure to the degradation of clear and dark vials were 41.96% and 36.91%[60].

Navel citrus fruits generally and gradually showed increasing chlorophyll a and b values, wherein the L*, b* and H * values gradually decrease during storage. During storage, the peel of the navel orange gradually changed from yellow-green to orange or orange-yellow. In addition, it was also found that there were no significant differences in the a * valuesin 3 ways during the storage period. The three storage methods were ventilating warehouse (VW), mechanical refrigeration warehouse (MRW), and mountain evaporate cooling ventilating warehouse (MECW)[61].

The stability of the tulip anthocyanin heating was sorted in red>pink≥orange-red>purple. The k value obtained indicated that the anthocyanins found in all samples of

tulips (red, orange-red, pink, purple) were more stable than the anthocyanins of black grapes. In addition, the value of $t_{1/2}$ red tulip anthocyanins was more stable than other tulip anthocyanins. The activation energy (Ea) of tulip anthocyanins ranged from 68.69-76 kj/mol. Studies have also shown that higher temperatures and longer heating times increase anthocyanin degradation in tulips[72] .

Table 2: Degradation of colors in food / beverage products

Products	Type of Testing	Results	Author(s)
Bayberry china wine	Temperature	The K value 25^{0}C was 0.0258, at 4^{0}C the k value was lower, namely 0.0030. The half-life value (t½) for the degradation of bayberry anthocyanins was 138.63 days at 4^{0}C, at 25^{0}C the half-life value (t½) was 26.87 days. The value of the activation energy (Ea) was 58.39 kj/mol.	Zhanget al [63]
Blueberry Juice, Blackberry Juice	Temperature	Anthocyanin degradation (64) from blueberries at any temperature followed the order kinetic model 1 reaction, half-life values (t½) at 40^{0}C, 50^{0}C, 60^{0}C, 70^{0}C and 80 ^{0}C were found to be 180.5, 42.3, 25.3, 86 and 5.1 hours. The activation energy (Ea) value of blackberry juice degradation during heating was 80.4kj/mol. Testing on blackberries (65) was carried out with a temperature range (100-180^{0}C) then divided into 2 sub ranges (100-140^{0}C and 140-180^{0}C). The activation energy (Ea) for NEB was from 100-180 ^{0}C (106kj/mol) slightly higher than that of anthocyanins at lower temperatures (92kj/mol). The value of the reaction rate constant at 140^{0}C for anthocyanin degradation ($3.5x10^{-3}$ s^{-1}) was reduced by 2 times than the NEB index ($1.6x10^{-3}$ s^{-1}) so that anthocyanin degradation was faster.	Kechinski et al [64], Jimenez et al [65]
Progmanate jam	Storage time and temperature	Jam stored at 5^{0}C. Results showed that 32% pigment degraded in HM and 14% reduced in LM after 150 days. All anthocyanin pigments showed reduction over time. Samples antosianindengan HM pectin had a value lower than the LM pectin at 5^{0}C.The a* value decreased at both storage temperatures. Light conditions did not have a significant effect on the color of the jam (a* value), the reduction was at a * = 47% and 51% on the bright day and dark after 150 days. HM progmanate jam was 35% more colorful than LM, where the a * value was reduced by 50% in both conditions	Melgarejo et al [66]
Strawberry juice	Temperature	Theresult of storage of unpasteurized strawberry juice had CIE L, a*, b* values of 35. 38, 37.49 and 20.46. The L and b* values of the sample were constant as a function of time regardless of storage temperature and bottle type. Meanwhile the value of a*decreased in all storage conditions. Some values decreased more rapidly during storage at higher temperatures in the bottle. This showed the effect of the storage temperature on the rate of red discoloration). The range of activation energy (Ea) values was 39-72 kj/mol (67). Storage of strawberry juice in refrigerators at 2-4^{0}C or room temperature of 30^{0}C showed no visual changes during the first 20 days. The values of L*, a*, b* and Ab_{510} indicated the stability of the refrigerator but have decreased at below room temperature. L*, a* and Ab_{510} stored at room temperature decreased by 43.8%, 34.88% and 14-28%. Results from Ab_{510} were more temperature sensitive than the L *, a*values,anthocyaninswhichdecreased at 25–30^{0}C (68).	Buve et al [67],Wang et al [68]
Orange juice	Temperature of 20^0, 28^0, 35^0 and 42^0 C for 32 weeks	The results of the study of orange juice showed a color change calculated using the CIELAB system and followed a 0 order reaction model. Before storage, the coordination of CIE L, a*, b* was 61.51, 3.20 and 54.55. All color parameters during storage changed significantly (p> 0.005). The decrease in L and b * values was seen in the orange juice turning black and the yellow color decreasing, but the a* value increased where the red color became more significant. Before storage C_{ab} and h_{ab}were 54.64 and 86.64. The activation energy (Ea) for all parameters ranged from 64 to 75 kj/mol (69). The thermal degradation of anthocyanins and visual color in orange juice followed 1st order reaction, and was associated with the Arrhenius model. The half-life value (t½) at 70^{0}C 2.5 was longer than 90 C. The activation energy (Ea) in anthocyanin degradation (55 kj/mol) was higher than the visual color reduction (47.51 kj/mol). The constant reaction values (k values) for visual color change were 0.0419, 0.0677, and 0.1049 at 70, 80 and 90 C, while the k valuesfor anyocyanin degradation were 0.0844, 0.1300, and 0.2487 at 70^0, 80^0 and 90^0 C (70) .	Wibowo et al[69], Shao-qian et al [70].

CONCLUSION

The research that has been conducted on the degradation of dyes originating from nature showed a decrease in the stability of these dyes. The decline in dye occurs due to external influences such as temperature, pH, light or sunlight, and storage time. The effect of temperature on the dye was indicated by the presence of dyes such as betacyanin, anthocyanin and carotenoids derived from natural dyes. These changes indicated a change in color, absorbance and a decrease or increase in degradation at too high a temperature. Too high a temperature will cause instability in natural dyes. The optimal temperature that has been investigated for betacyanin dye is below 70^{0}C. The effect of pH on the dye indicated a change in color and a decrease in degradation. The dyes examined at too low or too high pH result in dye breakdown. Therefore, it is best if the dye is in the normal pH range. The effect of light or sunlight on dyes causes changes that are almost the same as the effects of temperature and pH because sunlight contains ultraviolet light which can break down the dye, the longer exposure to sunlight will result in color changes.

Meanwhile, storage time causes the dye to become unstable, which is indicated by a change in color.

REFERENCES

1. Yusuf M, Shabbir M, & Mohammad F. Natural colorants: historical, processing and sustainable prospects, Nat Prod Bioprospect. 2017; 7:123-145.
2. Rymbai H, Sharma RR, & Srivastav M. Biocolorants and its implication in health anf food industry- a review. International Journal of PharmaTech Research, 2011; 3(4).
3. Sigurdson GT, Tang P, & Giusti MM. Natural colorants: food colorants from natural sources. Annual Revies of Food Science and Technology, 2017; 8:261-280.
4. Setiawan MAW, Nugroho EK., & Lestario LN. Ekstraksi betasianin dari kulit umbi bit (*Beta vulagaris*) sebagai pewarna alami. Jurnal Ilmu Pertanian, 2015; 27(1):38-43
5. Grzybowski A, & Majewski KK. What is color and how it is perceived? Clinic in Dermatology,2019; 37(5):392-401
6. Das M, Sacid A, Hossain, MDF, Jiang GH, Eun JB, & Ahmed M. Influence of extraction parameters and stability of betacyanins extracted from amaranth during storage. Journal Food Science and Technology, 2018; 56(2):643-653.
7. Visalakshi M, & Jawaharlal M. Healty hues-status and implication in industries- brief review. Journal of Agriculture and Allied Sciences, 2013; 3(2):42-51.
8. Maoka T. Carotenoids as natural functional pigments. Journal of Natural Medicines, 2019;7:1-16
9. Chew YM, Hung CH, & King VAE. Accelerated storage test of betalains extracted from peel of pitaya (*Hylocereus cacti*) fruit. Journal Food Science and Technology, 2019; 56:1595-1600.
10. Rahimi P, Abedimanesh A, Namin SAM, & Ostadrahimi A Betalains, the nature-inspired pigment, in health and diseases. Critical Reviews in Food Science and Nutrition, 2018; 59(1).
11. Jiang T, Mao Y, Sui L, Yang N, Li S, Zhenzhou Z, Wang C, Yin S, He J, & He Y. Degradation of anthocyanins and polymeric color formation during heat treatment of purple sweet potato extract at different pH. Food Chemistry, 2019; 274:460-470.
12. Akogou FUG, Kayode APP, Besten HMW, Linnermann AR, & Fogliano, V. Effects of processing and stroage on the stability of the red biocolorantsapigenidium from sorghum. LWT- Food Science and Technology, 2018; 90: 592-597.
13. Remini H, Mertz C, Belbahi A, Achir N, Dornire M, & Madani K. Degradation kinetic modelling of ascorbic acid and colour intensity in pasteurised blood orange juice durig storage. Food Chemistry, 2015; 173: 665-673.
14. Fenger JA, Moloney M, Robbins RJ, Collins TM, & Dangles O. The influence of acylation, metal binding and natural antioxidants on the thermal stability of red cabbage anthocyanins in neutral solutions. Food & Function, 2019; 10(10):6470-6751.
15. Lopez JAF, Angosto JM, Gimenez PJ, & Leon G. Thermal stability of selected red exctrats used as food colorants. Plant for Human Nutrition, 2013; 68:11-17.
16. Gimenez PJ, Lopez JAF, Angosto JM, & Obon JM., &Obon JM. Comparative thermal degradation patterns of natural yellow colorants used in foods. Plant Food for Human Nutrition, 2015; 70:380-387.
17. Argyropoulos D, & Muller J. Kinetics of change in colour and rosmarinic acid equivalents during concentive drying of lemon balm (*Melissa officinalis* L.). Journal of Applied Research on Medicinal and Aromatic Plants, 2014; 1(1):e15-e22.
18. Cisse M, Vailant F, Kane A, Nadiaya O, & Dornier M. Impact of the extraction procedure on the kinetics of anthocyanin and colour degradation of roselle extract during storage. Journal Science Food and Agriculture, 2011; 94: 1214-1221.
19. Guneser O. Pigment and color stability of beetroot betalains in cow milk during thermal treatment. Food Chemistry, 2016; 196: 220-277.
20. Pragalyaashree MM, Tiroutchelvame D, & Sashikumar S. Degradation kinetics of antocyanins extracted from roselle calyces (*Hibiscus sabdariffa*). Journal of Applied Pharmaceutical Science. 2018; 8(11):057-063.
21. Jaiswal AK, Gupta S, & Ghannam NA. Kinetic evaluation of colour, texture, polyphenols and antioxidant capacity of irishyork cabbage after blanching treatment. Food Chemistry, 2012; 131: 63-72.
22. Jaiswal AK., & Ghannam NA. Degradation kinetic modelling of color, texture, polyphenol and antioxidant capacity of york cabbage after microwave processing. Food Research International, 2013; 53:125-133.
23. Wibowo S, Grauwet T, Santiago JS, Tomic J, Vervoort L, Hendrikx M, & Loey AV. Quality changes of pasteurised orange juice during storage: a kinetic study of specific parameters and their relation to colour instability. Food Chemistry, 2015; 187: 140-151.
24. Idham Z, Muhammad II, & Sarmidi MR. Degradation kinetics and color stability of spray-dried encapsulated anthocyanins from Hibiscus sabdariffa L. Journal of Food Engineering, 2011; 35:522-542.
25. Cisilya T, Lestario LN, & Cahyanti MN. Kinetika degradasi serbuk antosianin daun miana (*Coleousscutellario des* L. Benth) Var. Crispa hasil mikroenkapsulasi. Chimica et Natura Acta, 2017; 5(3):146-152.
26. Patras A, Brunton NP, Donnell CO, & Tiwari BK. Effet of thermal processing on anthocyanin stability in foods; mechanism and kinetic degradation. Food Science & Technology, 2010; 21: 3-11.
27. Chandran J, Nishan P, Rekha SS, & Pandit AB. Degradation of colour in beetroot (*Beta Vulgaris* L.): a kinetic study. Journal Food Science and Technology, 2012; 51(10):2678-2684.
28. Prada AB, & Otero L. Effect of hyperbaric storage at room temperature on color degradation of strawberry juice. Journal of Food Engineering, 2016;109:141-143.
29. Santos CDd, Ismail M, Cassini AS, Marezak LDF, Tessaro IC, & Farid M. Effect of thermal and high pressure processing on stability of extracted from red beet stalks. Journal Food Science and Technology, 2017; 55(2):569-577.
30. Yusmita L, Kasim A, & Nurdi, H. Pengaruh suhu ekstraksi dan konsentrasi asam sitrat terhadap pigmen betasianin daun kremah merah (*Alternata dentata*) dan aplikasinya pada pangan. Pro Food (Jurnal Ilmu dan Teknologi Pangan), 2017; 3(1).
31. Patras A. Stability and colour evaluation of red cabbage waste hydroethanolic extract in presence of different food additivies or ingredients. Food Chemistry, 2018; 275:539-548.
32. Woo KK, Ngou FHN, Ago LS, Soong WK, & Tang PY. Stability of betalain pigment from red dragon fruit (*Hylocereuspolyrhizus*). American Journal of Food Technology, 2011;6(2):140-148.
33. Sari Y. Pengaruh pemanasan terhadap kestabilan pigmen betalain dari buah naga merah (Hylocereuspolyrhizus). Jurnal Pendidikan Kimia, 2018; 2(1).
34. Hidayat T, Pratjojo W, & Widiarti N. Uji stabilitas pigmen dan antioksidan ekstrak zat warna alami kulit buah naga. Indonesian Journal of Chemical Science, 2014; 3(2).
35. Khuzaimah S. Uji stabilitas pigmen hasil ekstraksi zat warna alami dari kulit buah naga (Hylocereusundatus). JTI, 2018; 2(2).
36. Kwartinigsih E, Prastika AK, & Triana DL. Ekstraksi dan uji stabilitas antosianin dari kulit buah naga merah super (Hylocereuscostaricensis). Prosiding, 2016
37. Sarkis JR, Jaeschke DP, Mercali GD, Tessaro LC, & Marczak LDF. Degradation kinetic of anthocyanins in blackberry pulp during ohmic and conventional heating. International Food Reasearch Journal, 2019; 26(1):87-97.
38. Sipahli S, Mohanlall V, & Mellem JJ. Stability and degradation kinetic of crude antocyanins extracts from H. sabdariffa. Food Science and Technology, 2017; 37(2):209-215.
39. Wu HY, Yang KM, & Chiang PY. Roselle anthocyanins: antioxidant porperties and stability to heat and pH. Molecules, 2018; 23(1357).

40. Fendri STJ, Martinus BA, & Haryanti MD. Pengaruh pH dan suhu terhadapstabilitas antosianin dari ekstrak kulit ubi jalar ungu (Ipomoea Batatas (L.) Lam). Chempublish Journal, 2018; 2(2).
41. Jie L, Xiao-ding L, Yun Z, Zheng-dong Z, Zhi-ya Q, Meng L, Shao-hua Z, Liu S, Meng W, & Lu Q. Identification and thermal stability of purple-fleshed sweet potato anthocyanins in aqueous solutions with various pH values and fruit juices. Food Chemistry, 2013; 136:1429-1434.
42. Mulyawanti I, Budijanto S, &Yasni S. Stability of anthocyanins during processing, storage and stimulated digestion of purple sweet potato pasta. Indonesian Journal of Agricultural Science, 2018; 19(1):1-8.
43. Herrero JAH, & Frutos MJ. Degradation kinetics of pigment, colour and stability of the antioxidant capacity in juice model system from six anthocyanin sources. International Journal of Food Science and Technology, 2011; 46:2550-2557.
44. Vital DL, Li Q, West L, & Meija EGd. Anthocyanin condensed forms do not affect color or chemical stability of purple corn pericarp extract stored under different pH. Food Chemistry, 2017; 232:639-647.
45. Goncalves EM, Abreu M, Brandio TSR, & Silva CLM. Degradation kinetics of colour, vitamin C and drip loss in frozen brocolli (Brassica oleracea L. ssp. Italica) during storage at isothermal and non-isothermal conditions. International Journal of Refrigeration, 2011; 34:2136-2144.
46. Musche BM, Speers RA, & Ruspasinghe HRV. Storage temperature impacts on anthocyanina degradation, color changes dan haze development in juice of "Merlot" and "Ruby" grapes (*Vitis vinifera*). Frontier in nutrition, 2012;5(100).
47. Moldova B, David L, Chishora C, & Cimpoiu C. Degradation kinetics of anthocyanins ferom European Cranberrybush (*Viburnum opulus* L.) fruit extracts, effests of temperature, pH and storage. Mocules, 2012; 17:11655-11666.
48. Bastante MJC, Hurtando N, Delgado A, & Heredia FJ. Impact of pH and temperature on the colour and betalain content of colombian yellow pitaya peel (*Selenicereus Megalanthus*). Journal Food Science and Technology, 2016; 53(5):2405-2413.
49. Ali MA, Yusof YA, Chin NI, Ibrahim MN, & Basra SMA. Drying kinetics and colour analysis of Moringa Oleifera leaves. Agriculture and Agricultural Science Procedia, 2014; 2:394-400.
50. Chunthawon S, Achariyaviriya S, Achariyaviriya A, & Nmasanguam K. Color kinetics of longan flesh drying at high temperature. Procedia Engineering, 2012; 32:104-111.
51. Mercali GD, Schwartz S, Marezak LDF, Tessaro IC, & Sastry S. Effect of the electric field frequency on ascorbic acid degradation during thermal treatment by ohmic heating. Journal Agric Food Journal Agric Food Chem, 2013; 65(25):5865-5870.
52. Sonawane A, Pathak S, & Pradhan RC. Effect of processing temperature on dynamic rheological properties and color degradation kinetics of bael fruit pulp. Journal on the Science of Food and Agriculture, 2020.
53. Ahmed J, Al-Salman F, & Almusallam AS. Effect of blanching on thermal color degradation kinetics and rheological behavior of rocket (*Eruca sativa*) puree. Journal of Food Engineering, 2013; 119:660-669.
54. Wawire M, Oey I, Mathooko FM, Njoroge CK, Shitanda D, & Hendrickx M. Kinetic of thermal inactivation of peroxidase and color degradation of African Cowpea (*Vigna unguiculata*) leaves. Journal of Food Science, 2016; 91(1).
55. Fratianni A, Albanese D, Mignogna R, Luciana C, Panfili G, & Matten MD Degradation of carotenoids in Apricot (*Prunus armeniaca* L.) during drying process. Plant Foods for Human Nutrition, 2013; 68(3):241-246.
56. Kara S, & Ercelebi EA. (2013). Thermal degradation kinetics of anthocyanins and visual colour of Ummu mulberry (*Morus nigra L.*). Journal of Food Engineering, 2013; 116:541-547.
57. Lixia H, Ling B, & Shaojin W. Kinetic of color degradation of chesnut kernel during thermal treatment and storage. International Journal Agriculture & Biological Eng, 2015; 8(4).
58. Rengku PM, Ridhay A, & Prismawiryanti. Ekstraksi dan uji stabilitas betasianin dalam ekstrak buah kaktus (*Opuntia elatior* Mill.). Kovalen, 2017; 3(2):142-149.
59. Kurniati N, Prasetya AT, & Winarn. Ekstraksi dan uji stabilitas zat warna brazilien dari kayu secang (*Caesalpinia sappan* L.,). Indonesian Journal of Chemical Science, 2012; 1(1).
60. Rundubello BA, Ridhay A, Hardi J, & Puspitasari DJ. Uji stabilitas pigmen esktrak ubi banggai (*Dioscoreabulbifera* var celebica Burkill) pada berbagai variasi pH dan lama paparan sinar matahari. KOVALEN: Jurnal Riset Kimia, 2019; 5(1):9-16.
61. Gao Y, Liu Y, Kan C, Chen M, & Chen J. Changes of peel color and fruit quality in navel orange fruits under different storage methods. Scientia Horticultura, 2019;256:108522.
62. Sagdic O, Ekici L, Ozturk I, Tekinay T, Polat B, Tastemur B, Bayram O, & Sentruck B. Cytotoxic and bioactive properties of different color tulip flowers and degradation kinetic of tulip flower anthocyanins. Food and Chemical Technology, 2013; 58:432-439.
63. Zhang Z, Li J, & Fan L. Evaluation of the composition of Chinese bayberry wine and its effects on the color changes during storage. Food Chemistry, 2018; 276:451-457.
64. Kechinski CP, Guimaraes PVR, Norena CPZ, Tessaro IC, & Marozak LDF. Degradatiom kinetics of anthocyanins in blueberry juice during thermal treatment. Journal of Food Science, 2010; 75(2): C173-176.
65. Jimenez N, Bohuon P, Lima J, Dornier M, Vailnat F, & Perez AM. Kinetics of anthocyanins degradation and browning in reconstituted blackberry juice treated at high temperaturs (100-180 C). Journal of Agricultural and Food Chemistry, 2010; 58:2314-2322.
66. Melgarejo P, Martinez R, Hernandez F, Martinez JJ, & Legua P.. Anthocyanin content and color development promagnate jam. Food and Bioproduct Technology, 2011; 89:477-481.
67. Buve C, Kebede BT, Batseller CD, Carrilo C, Pham HTT, Hendrick M, Grauwet T, & Loey AV. Kinetics of colour changes in pasteurised strawberry juice during storage. Journal of Food Engineering, 2017; 216:42-51.
68. Wang Z, Zhang M, & Wu Q. Effects of temperature, pH, and sunlight exposure on the color stability of strawberry juice during processing and storage. LWT-Food Science and Technology, 2014; 60(2):1174-1178.
69. Wibowo S, Vervoort L, Tonic J, Santiago JS, Lemmens L, Pannozo A, Grauwet T, Hendrikx M, & Loey A. Colour and carotenoid changes of pasteurised orange juice during storage. Food Chemistry, 2015; 171; 330-340.
70. Shao-qian C, Luang L, & Si-yi P. Thermal degradation kinetics of anthocyanins and visual color of blood orange juice. Agricultural Science, 2011; 10(12):1992-199.