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Tuning the stability of red color natural pigments in fruit extracts by pH control

Leny Yuliati^{1,2*}, Juliana² and R Indrawati^{1,2}

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¹Ma Chung Research Center for Photosynthetic Pigments, Universitas Ma Chung, Villa Puncak Tidar N-01, Malang 65151, East Java, Indonesia

²Department of Chemistry, Faculty of Science and Technology, Universitas Ma Chung, Villa Puncak Tidar N-01, Malang 65151, East Java, Indonesia

Email : * leny.yuliati@machung.ac.id

Abstract. Natural pigments have been recognized as potential sources for natural colorants and visible light absorbers. Unfortunately, most natural pigments have problems with the stability, which limited their potential uses for various applications. In this study, extraction of five types of red fruits, which were tomato, watermelon, strawberry, carrot, and papaya, was carried out by a slow juicer. After drying by a freeze dryer, the stability of each extract was then monitored by color measurements at various pH of 1 –10 for 7 days. The results showed that the stability of the pigments was affected by the pH. However, most of the pigment extracts still suffered from the stability issue as the red color scales were found to decrease with the increase of the storage time in most any pH conditions. Particular attention was made to the strawberry extract that could be successfully stabilized at pH of 1. In addition to its stability, the strawberry extract also gave the highest red color scale among all the investigated extracts. This result suggested that the stability of the strawberry extract was tunable by the pH control and thus, its potential use as a red color source for various applications could be expected.

1. Introduction

Growing interest in the use of red color natural pigments have been nurtured by the facts that natural pigments are non -toxic and good for health [1,2] and therefore, shall be used to replace the artificial coloring in food industry [3,4]. Aside of the food industry, the possibility to utilize the natural pigments for various other applications is still investigated and considered as one of the on-going important research topics. One potential use of the natural pigment is as the light harvester to be applied in photonic materials such as light emitting diodes, photocatalysts, and photovoltaics. However, such natural pigments shall have good stability, which is unfortunately not the original feature of the natural pigments. Recently, special attentions have been made to produce the semiconductor materials with red color property for photocatalytic applications [5-8]. These red color materials are expected to be able to harvest the full spectrum of visible light from the solar so that better efficiency in the solar energy conversion could be realized.

It has been reported that the natural pigments responsible for the red color in plants are mostly lycopene, anthocyanins and betacyanins [9,10]. The group of phytochemicals for lycopene, anthocyanins, and betacyanins are carotenoid, flavonoid, and betalains, respectively. Lycopenes can be obtained from various fruit sources, such as tomato, watermelon, and papaya [11,12]. On the other

hand, the anthocyanins could be taken from strawberry [13]. Another pigment that is responsible to give the red-orange color such as in carrot is α -carotene and β -carotene [11,14].

In order to be applicable as natural colorant and light harvester, the stability of the natural pigments shall be highly considered. Unfortunately, most of these red pigments are unstable and it still remains as the crucial issue to be solved. In this study, the stability of red pigments obtained from five red fruits, which were tomato, watermelon, strawberry, carrot, and papaya, were investigated under various pH conditions for several days. This study demonstrated that the stability of the fruit extract could be affected by the pH. Among the investigated fruit extracts, the strawberry extract showed the most intense red color and the most stable one under acid condition.

2. Experimental

2.1. Extraction of red fruits

Red fruits used in this study were tomato, watermelon, strawberry, carrot, and papaya, which were commercially purchased from a fresh fruit market in Malang, East Java, Indonesia. These fresh fruits were cleaned thoroughly by using a flow water. After cutting them into small pieces, the fruits were then squeezed by a HUROM slow juicer (HH-SBF11) without addition of any water. The extracts were then collected and weighed. The moisture content of the fresh pigment extracts was measured by a moisture analyzer (Shimadzu MOC63u).

2.2. Encapsulation and drying process

The fruit extracts were encapsulated by using a maltodextrin (Yishui Dadi Corn Developing Co., Ltd., dextrose equivalent of 10–12%), where the ratio of the total mass of maltodextrin and fruit extract to the total volume of water and fruit extract was fixed to 0.1–0.2%. A certain amount of maltodextrin was dissolved in a total of 100 ml of water at 333 K. The maltodextrin solution was then added into the fruit extract and homogenized by a homogenizer to get a well-mixed solution. The obtained mixture was then put into a freezer for overnight, followed by a drying process using a freeze dryer for 48 h at 228 K. The pressure during the freeze drying process was set at 0.02 MPa. The obtained dried pigment was crushed in a mortar and kept in a bottle. After nitrogen gas flow, the bottle was closed tight and sealed. The dried pigment was then kept in the freezer at low temperature of 258 K. The moisture content of the dried pigment extracts was measured by a moisture analyzer (Shimadzu MOC63u).

2.3. Preparation of buffer solution

In order to prepare buffer solutions, various chemicals were purchased and used as received. The chemicals were potassium chloride (KCl, Merck, 99%), chloride acid (HCl, Merck, 37%), citric acid ($C_6H_8O_7$, Chameleon, 99%), potassium dihydrogen phosphate (KH_2PO_4 , Chameleon, 99%), sodium tetraborate decahydrate ($Na_2B_4O_7 \cdot 10H_2O$, Wako, 99%), trisodium citrate ($Na_3C_6H_5O_7$, Chameleon, 99%), and sodium hydroxide (NaOH, Chameleon, 96%). The buffer solutions with pH of 1–10 were prepared in the similar way as the literature [15], by first mixing the solution A and B as shown in the Table 1. The obtained solution was then diluted to get 100 mL of buffer solution.

Table 1. List of chemicals required to prepare buffer solutions with various pH

Buffer pH	Solution A	Solution B
11	0.2 M KCl (25 mL)	0.2 M HCl (48.5 mL)
2	0.2 M KCl (25 mL)	0.2 M HCl (6.5 mL)
3	0.1 M $C_6H_8O_7$ (46.5 mL)	0.1 M $NaC_6H_7O_7$ (3.5 mL)
4	0.1 M $C_6H_8O_7$ (33 mL)	0.1 M $NaC_6H_7O_7$ (17 mL)
6	0.1 M $C_6H_8O_7$ (20.5 mL)	0.1 M $NaC_6H_7O_7$ (27 mL)
6	0.1 M KH_2PO_4 (50 mL)	0.1 M NaOH (5.6 mL)
7	0.1 M KH_2PO_4 (50 mL)	0.1 M NaOH (29.1 mL)

8	0.1 M KH ₂ PO ₄ (50 mL)	0.1 M NaOH (46.1 mL)
9	25 mM Borax (50 mL)	0.1 M HCl (4.6 mL)
10	25 mM Borax (50 mL)	0.1 M NaOH (18.3 mL)

2.4. Stability test

The effect of pH on the stability of the pigment extract was studied by measurements of color changes obtained on each extract. In a typical test, 0.375 g of the extract was dissolved in 25 mL of each buffer solution. The solution was divided into five glass bottles so that the content of each bottle was 5 mL. The stability test for each pigment was then carried out under the dark condition for 7 days in a climate chamber where the temperature and the relative humidity (RH) were maintained at 298 K and 15%, respectively. The color measurement for each extract was conducted at 0, 1, 2, 4, and 7 days by using a ColorFlex EZ spectrophotometer (HunterLab), where the wavelength was measured from 400 to 700 nm with an interval of 10 nm. The results were shown as luminance or lightness (L*), redness (a*) and yellowness (b*), following the L*a*b* colorimetric model (CIELab) developed by the International Commission on Illumination (Commission Internationale de l'Éclairage, CIE). The values shown were the average values taken from three repetitions.

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


3. Results and Discussion



3.1. Physical properties of fruit extracts

The water contents of all the fresh and dried fruit extracts are listed in Table 2. It was obtained that the fresh extracts have high water content of ca. 85–95%. Among the investigated extracts, the tomato extract has the highest water content (Entry 1), followed by watermelon, strawberry and carrot extracts (Entries 2-4). The papaya extract showed the least water content (Entry 5). As expected, after the drying process using the freeze dryer, the water content obtained on the extracts could be significantly reduced from ca. 85–95% to ca. 3–5%. This result showed that the drying process employed here was good enough to produce the dried pigment extracts.

It was worthy to note that all the dried extracts could maintain their pigment colors, which were red and red-orange colors. This is in good agreement with previous claims that the freeze drying method is a good approach to reduce the degradation of the pigment [16,17]. These red and red-orange colors could be compared by measuring the redness (a*) and yellowness (b*) of the extracts, which would be discussed in section 3.2. Judging from the color of the dried extract, the strawberry extract showed the most intense red color (Table 1, Entry 3), while the other extracts gave less red and more red-orange color. Therefore, among all the extracts, the strawberry extract might be the most suitable one as the red pigment source.

Table 2. Water contents in fresh and dried pigment extracts and their physical color appearances

Entry	Pigment Source	Water Content (%)		Color Appearance
		Fresh	Dried	
1	Tomato	94.57	4.71	
2	Watermelon	93.89	3.18	
3	Strawberry	92.92	5.10	

4	Carrot	91.37	4.57	
5	Papaya	84.85	5.50	

3.2. Effect of pH

In order to study the effect of pH on the stability of pigment extracts, the color measurement of each pigment extract was performed at different pH in the range of 1–10. The measurement was carried out on the same day without any storage time. As shown in Figure 1(a), there was only a slight change in the lightness (L^*), redness (a^*), and yellowness (b^*) of tomato extract with the changes of the pH. No clear trend could be observed when the solution pH was varied. This result was in good agreement with the color appearance of the solution. Shown in Figure 1(b) is the similar color of tomato extracts in various solution pH.

The effect of pH on the L^* , a^* , and b^* values of the watermelon extract is shown in Figure 2(a). It was also observed that the variation of pH did not give much influence on the color changes in the watermelon extract. The lightness values were found to be slightly dropped from 70.5 to 66.7 when the pH solution was 6–8. On the other hand, the redness and yellowness improved from 1.52 to 2.09, and from 3.75 to 4.81, respectively, at pH of 6–8. This result showed that the watermelon extract might be better stabilized in such pH range. The color appearance of the watermelon extract in various solution pH could be seen in Figure 2(b).

Figure 3 shows the effect of pH on the color scales of strawberry extracts measured in pH 1–10 and their color appearances. Different from the case of tomato and watermelon extracts, there was a clear change in the color of the strawberry extract with the change of the pH. It could be observed that the strawberry extract showed the highest redness and yellowness when the solution was acidic with pH of 1 and 2. The values decreased with the increase of the pH from 3 to 5 and reach almost constant values up to pH of 9. At pH of 10, the yellowness did not much change, but the redness level increased. However, the increase was not as much as those in pH 1 and 2. This result suggested that the color intensity of the strawberry extract could be easily tuned by the pH.

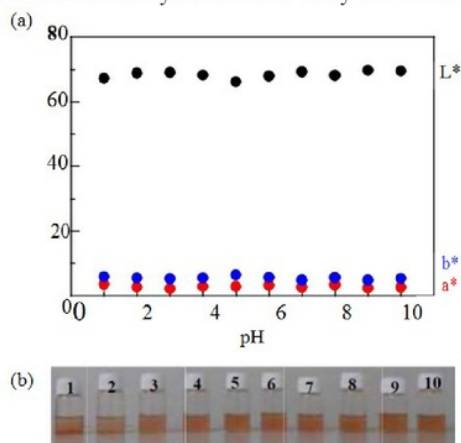


Figure 1. (a) Effect of pH on the values of L^* , a^* , and b^* of tomato extracts and (b) their respective color appearances.

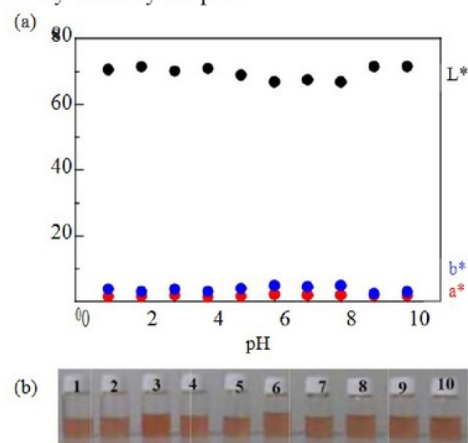


Figure 2. (a) Effect of pH on the values of L^* , a^* , and b^* of watermelon extracts and (b) their respective color appearances.

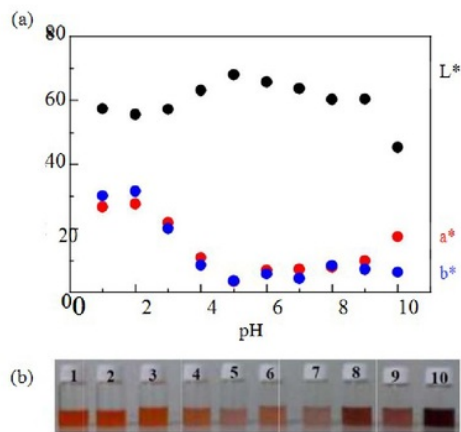


Figure 3. (a) Effect of pH on the values of L^* , a^* , and b^* of strawberry extracts and (b) their respective color appearances.

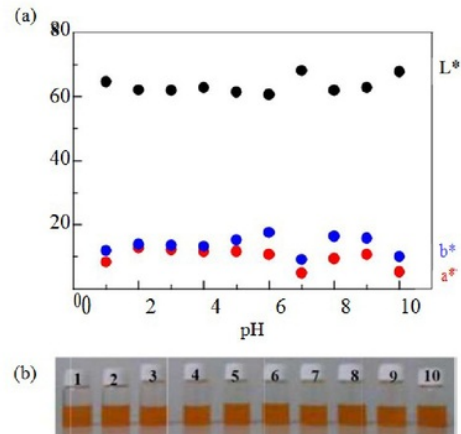


Figure 4. (a) Effect of pH on the values of L^* , a^* , and b^* of carrot extracts and (b) their respective color appearances.

The effect of pH for carrot extract is shown in Figure 4. The color scales of the carrot extract was nearly unchanged when the pH was at 1–6. However, the clear drop of the color scales could be observed when the solution was neutral at pH 7, which resulted in the increase of the lightness. The a^* and b^* values increased again with the solution pH of 8 and 9, and reached low values at pH of 10. While the trend was not clear, the changes in the color scales in the carrot extracts were not as obvious as those observed in the strawberry extracts. As a result, there was no clear change in the physical appearance of the extracts, as shown in the Figure 4(b). It was obtained that the carrot extracts were red-orange in color, in good agreement with their levels of redness and yellowness.

Figure 5 shows the changes of L^* , a^* , b^* , and the color appearance of the papaya extract with the changes in the solution pH. Similar to carrot extract, papaya extract also showed higher scale of yellowness than the redness. However, no clear color change occurred with the change of the pH. All the lightness, redness, and yellowness were maintained to be similar in all pH conditions.

Based on all the results mentioned above, it was obtained that the color appearances of the fruit extracts were not much affected by the pH, except for the strawberry extract. The differences observed here shall be strongly related to the different types of major natural pigment in the fruit extract. As has been reported, anthocyanins would be the responsible pigment giving the red color in strawberry [1]. This study suggested that the pH affected the color of the anthocyanins in the strawberry extract.

3.3. Effect of storage time

Effect of storage time was further studied for each extract with various pH of 1–10 after 1, 2, 4, and 7 days. Particular attention was made on the redness scale (a^*) to evaluate the stability of the extract over the storage time. Displayed in Figure 6 is the changes in the values of a^* after the tomato extracts at different pH were kept in the climate chamber for 1, 2, 4, and 7 days. In all pH conditions, the a^* values dropped after the extracts were kept for several days. For instance, at the initial condition the highest a^* value observed on tomato extract was 3.55 at pH 1, but the number was kept decreasing within the time and it reached the value of 1.93 after 7 days. Similar results could be observed for other solutions at different pH, suggesting that the tomato extract was not stable and easily degraded.

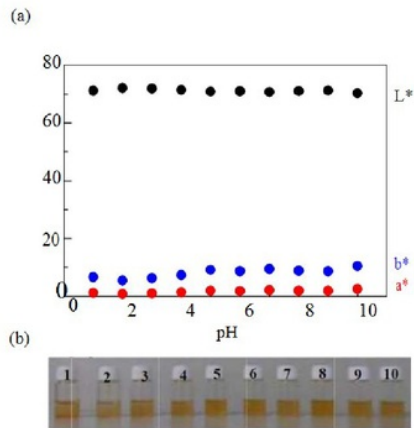


Figure 5. (a) Effect of pH on the values of L*, a*, and b* of papaya extracts and (b) their respective color appearances.

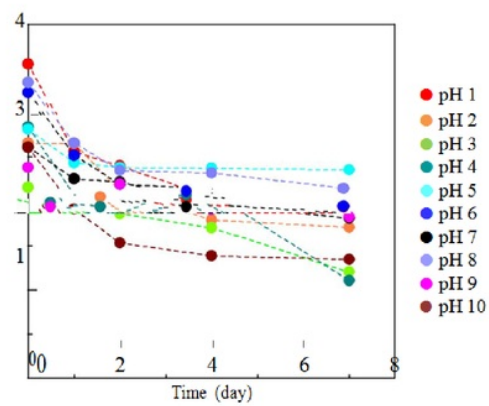


Figure 6. Effect of storage time on the a* values for tomato extracts prepared in different solutions with pH of 1-10.

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Figure 7 shows the effect of storage time on the stability of the watermelon extracts. Similar to those observed in the tomato extracts, the watermelon extracts also could not stand with the degradation issue. The a* values decreased within the time and even worse than the tomato extracts. As an example, the highest a* value was 2.09 at pH 6, but it decreased drastically to 0.46 within 7 days. An extreme result could be also observed when the watermelon extract was kept under pH of 10. The a* value decreased from 1.77 to -0.15 after 7 days. The negative value showed the greenness in the color scale. This result clearly showed that the watermelon extract was also unstable under all pH conditions.

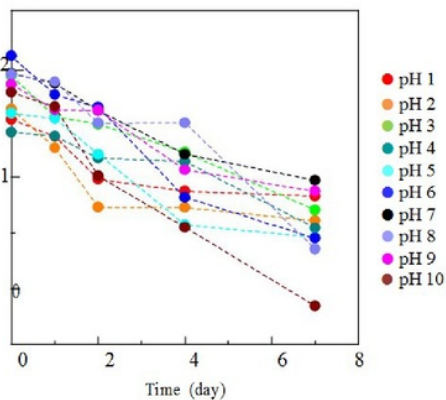


Figure 7. Effect of storage time on the a* values for watermelon extracts prepared in different solutions with pH of 1-10.

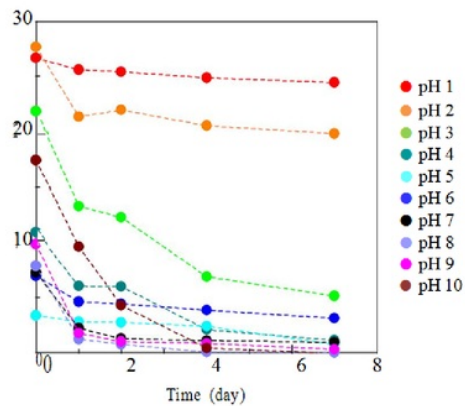


Figure 8. Effect of storage time on the a* values for strawberry extracts prepared in different solutions with a pH of 1-10.

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The stability of the strawberry extract was also investigated and the results are shown in Figure 8. As also displayed in Figure 3, strawberry extracts have the most intense red color that could be expressed by their high a* values. In contrast to the tomato and the watermelon extracts, the strawberry extract showed good stability when the solution was in acid condition, such as at pH 1. The a* value at pH 1 was initially at 26.7 and it remained high (24.4) after 7 days. Such low change in the a* value could be considered as a good indicator of the stability of the strawberry extract. While the stability

could be maintained at low pH, it could not be achieved when the pH solution increased. When the pH was 10, the a^* value even dropped to a negative value (-0.06), which could be referred to the green color, suggesting that the red pigment in the strawberry extract was totally diminished at high pH after 7 days. These results showed that for the strawberry extract, pH 1 played as important parameter to maintain not only the red color but also its stability.

Figure 9 displays the changes in the a^* values for carrot extracts dispersed in solutions with different pH after 1, 2, 4, and 7 days. Carrot extract also suffered from the stability issue. As shown in Figure 9, the highest a^* value of 12.7 was obtained when the pH solution was 2. Even though it seemed that the carrot extract could be stabilized for the first 4 days, eventually the a^* value decreased to 5.1 after 7 days. This result clearly suggested the instability of the carrot extracts under the current conditions.

The redness level in papaya extracts was also decayed with the increase of the storage time. As shown in Figure 10, the decrease in the a^* values could be observed in the papaya extracts under all pH conditions. Even though the a^* value was near to constant after 2, 4, and 7 days at pH 8, it indeed decreased to more than 50% off its initial a^* value, which was from 1.89 to 0.93. Based on these results, it could be suggested that the papaya extracts were not that stable to be used as the red pigment source.

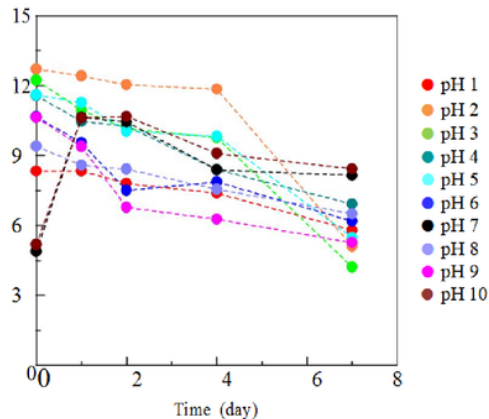


Figure 9. Effect of storage time on the a^* values for carrot extracts prepared in different solutions with pH of 1–10.

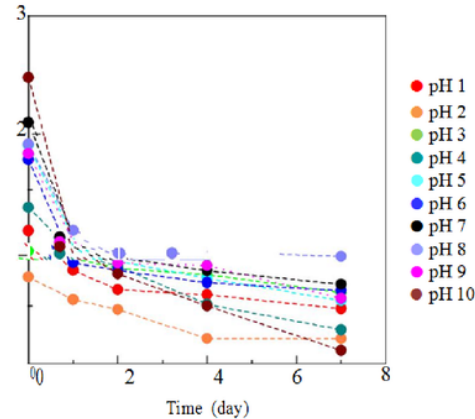


Figure 10. Effect of storage time on the a^* values for papaya extracts prepared in different solutions with pH of 1–10.

4. Conclusions

The stability of the pigment extracts from five fruits, which were tomato, watermelon, strawberry, carrot, and papaya, in various pH for 1, 2, 4, and 7 days was evaluated by color measurements. Among all the investigated extracts, the strawberry extract gave the highest redness value (a^*) of *ca.* 27. Furthermore, only strawberry extracts showed good stability even after 7 days when the solution was acidic with pH of 1. The loss of the a^* value was less than 9% after 7 days, suggesting the good stability of the strawberry extract as compared to other investigated fruit extracts. This study demonstrated that the use of the strawberry extract shall be enlarged for various applications by employing the required conditions to maintain the stability of the red pigment in the strawberry, such as the acidic condition.

2 Acknowledgement

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References

- [1] He J and Giusti M M 2010 *Annu. Rev. Food Sci. Technol.* **1** 163
- [2] Wootton-Beard P C and Ryan L 2011 *Food Res Int.* **44** 3135
- [3] Wrostand R E and Culver C A 2012 *Annu. Rev. Food Sci. Technol.* **3** 59
- [4] Sigurdson G T, Tang P and Giusti M M 2017 *Annu. Rev. Food Sci. Technol.* **8** 261
- [5] Xu X, Randorn C, Efstathiou P and Irvine J T S 2012 *Nat. Mater.* **11** 595
- [6] Liu G, Yin L-C, Wang J, Niu P, Zhen C, Xie Y and Cheng H-M 2012 *Energy Environ. Sci.* **5** 9603
- [7] Yu Y-G, Chen G, Yang X-K, Zhou Y-S and Han Z-H 2014 *Int. J. Hydrogen Energ.* **39** 13534
- [8] Cai, B, Wang, J, Gan S, Han D, Wu Z and Niu L 2014 *J. Mater. Chem A* **2** 5280
- [9] Delgado-Vargas F, Jiménez A R and Paredes-López O 2000 *Crit. Rev. Food Sci. Nutr.* **40** 173
- [10] Leong H Y, Show P L, Lim M H, Ooi C W and Ling T C 2018 *Food Rev. Int.* **34** 463
- [11] Khoo H-E, Prasad K N, Kong K-W, Jiang Y and Ismail A 2011 *Molecules* **16** 1710
- [12] Perveen, R, Suleria H A R, Anjum F M, Butt M S, Pasha I and Ahmad S 2015 *Crit. Rev. Food Sci. Nutr.* **55** 919
- [13] Aberoumand A 2011 *World J. Dairy Food Sci.* **6** 71
- [14] Rao A V and Rao L G 2007 *Pharmacol. Res.* **55** 207
- [15] Gomori, G. 1955 Preparation of buffers for use in enzyme studies *Preparation and Assay of Enzymes*, Vol 1, ed N. Kaplan and N. Colowick (Cambridge: Academic Press, Inc.) chapter **16** 138–146
- [16] Desobry, S A, Netto F M and Labuza T P 1997 *J. Food Sci.* **62** 1158
- [17] Tang Y C and Chen B H 2000 *Food Chem.* **69** 11

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