ELSEVIER

Contents lists available at ScienceDirect

Chinese Journal of Analytical Chemistry

journal homepage: www.elsevier.com/locate/cjac



A portable colorimetric tool using a smartphone camera applied for determining total phenolic contents in coffee products

Le-Thi ANH-DAO ^{a,b}, Nguyen THANH-NHO ^a, Bui HUU-TRUNG ^c, Nguyen TIEN-GIANG ^c, Thach UT DONG ^d, Nguyen QUOC-DUY ^a, Nguyen QUANG-HIEU ^a, Nguyen LE-VY ^a, Nguyen-Thi THANH-DIEU ^a, Dien Vu Thanh TO ^{e,f}, Do MINH-HUY ^{a,*}, Nguyen CONG-HAU ^{a,*}

- ^a Faculty of Environmental and Food Engineering, Nguyen Tat Thanh University, Ho Chi Minh City 70000, Vietnam
- ^b School of Earth Sciences and Environmental Engineering, Gwangju Institute of Science and Technology (GIST), Gwangju 61005, Korea
- Faculty of Chemical and Food Technology, Ho Chi Minh City University of Technology and Education, Ho Chi Minh City 70000, Vietnam
- ^d Faculty of Pharmacy, Ton Duc Thang University, Ho Chi Minh City, Vietnam
- ^e Institute for Advanced Material Technology, Van Lang University, Ho Chi Minh City 700000, Vietnam

ARTICLE INFO

Keywords: Portable colorimetric tool Total phenolic contents Coffee Smartphone Lam Dong

ABSTRACT

In the present study, a self-designed colorimetric box equipped with a smartphone as an emerging analytical approach was designed to determine the total phenolic contents (TPCs) in coffee. The formed blue complex between the phenolic compounds and Folin-Ciocalteu reagent was measured its color intensities using RGB (red, green, blue) color system. Different parameters related to the photo capturing were investigated, i.e., the distance between the smartphone and sample holder, constant illumination inside the box, and the reaction time. The method performance based on three color channels (R, G, and B) was evaluated and agreed with Appendix F of AOAC (2016). Different sensitivities among three color channels were achieved, and the linear ranges were wide, from 10.0 mg GAE L⁻¹ up to 120.0 mg GAE L⁻¹ for the B channel, which was compatible with various coffee types (GAE: gallic acid equivalent). No statistical difference between the smartphone-based methods and standard Ultraviolet-visible (UV-Vis) was observed by t-test (P = 95%). Various advantages were obtained, e.g., simplicity, portability, low-cost operation, and easy accessibility for producers and consumers to test for the raw materials, intermediate and final products. The proposed method was applied to several specific roasted ground coffee products from Lam Dong Province (Vietnam) in the context of the limited scientific data on the internal composition of Vietnamese coffee. The results demonstrate that the TPCs depend on coffee varieties, i.e., Robusta exhibits higher TPCs than Arabica, and roasting levels during the processing, i.e., dark mode generally performs the lowest TPCs, due to the thermal decomposition and formation of different phenolic compounds by Millard reactions.

1. Introduction

Coffee is among the most frequently consumed beverages around the world, thanks to the presence of pleasant taste, aroma, and stimulating properties [1,2]. It could promote human health by providing many biologically active compounds (chlorogenic acid, diterpenes, nicotinic acid, tannic acid, trigonelline, pyrogallic acid, and especially caffeine) [1,3]. These compounds are claimed to associate with decreased risks of cardiovascular diseases, type 2 diabetes, mortality, and several cancers, such as breast, prostate, liver, and endometrial cancers [4]. *Coffea arabica* (Arabica) and *Coffea canephora* (Robusta) are the most commercially important species grown worldwide. Commercially, arabica cof-

fee is responsible for around 70 percent of the global coffee market, and robusta coffee accounts for the rest [5,6].

Coffee quality strongly depends on the roasting levels of the beans, which affects the drink's flavor, aroma, and color [7]. Roasting is a complicated process involving the careful application of heat to green coffee for a set amount of time, depending on the desired characteristics of the final product [8]. During the roasting process, various reactions potentially occur, causing substantial changes in chemical composition and sensory characteristics [9]. For example, green coffee beans contain about 200 aroma compounds, while around 1000 have been identified in roasted beans [10], or the increase in roasting time of coffee beans can generate higher intensities of bitter and burnt characteristics [11].

E-mail addresses: thachutdong@tdtu.edu.vn (T. UT DONG), dmhuy@ntt.edu.vn (D. MINH-HUY), nchau@ntt.edu.vn (N. CONG-HAU).

^f Faculty of Applied Technology, School of Engineering and Technology, Van Lang University, Ho Chi Minh City, Vietnam

^{*} Corresponding authors.

Caffeine and other phenolic compounds or polyphenols, such as chlorogenic acid, diterpenes, and trigonelline, are important and significantly contribute to coffee beverages' sensory and antioxidant properties [12]. However, the phenolic compounds in coffee are heavily influenced by roasting, i.e., light, medium, medium-dark, and dark roasting degrees, typically due to changes in temperature and time [11]. It is, therefore, essential to determine the total phenolic contents (TPCs) in different coffee varieties to assess their content to find a correlation between the types and roasting degrees and the content of phenolic compounds or polyphenols.

In the literature, high-performance liquid chromatography with ultraviolet-visible (UV-Vis) detector (HPLC-UV/Vis) and ultra-high performance liquid chromatography coupled with (tandem) mass spectrometry (UHPLC-(MS/) MS) are well-established techniques to determine phenolic compounds and polyphenols in coffee [6,13,14]. Despite the great advantages of these instruments, most of these methods are limited by being time-consuming, laborious, demanding a relatively large amount of reagents, and high cost of devices, so these methods are inapplicable for in situ measurements. The method based on UV-Vis spectrophotometry using Folin-Ciocalteu can be used as a measurement standard for determining the TPCs in coffee [15-18]. Although the method is accurate, specific instruments and trained operators in laboratories are also required, which would limit the users, e.g., untrained producers, customers, etc., as well as the application context, e.g., on-site measurement, quick tests. To address these limitations, a rapid, simple, costefficient, and promising portable field use approach using digital image colorimetry based on a smartphone has recently emerged as an alternative to conventional methods due to the smartphone's popularity in society. Smartphone digital image colorimetry is based on the conversion of the pixel intensity of images captured for the sample by a smartphone camera to its three primary color channels, red-green-blue (RGB) components allocated of every primary color ranging from 0 to 255 for entirely black and white, respectively [19]. Smartphone-based digital image colorimetry has been applied for determining a wide range of analytes, including TPCs in tea [20], TPCs in beer [21], copper in edible oil samples [22], Fe (III) in water [23], carbaryl in food samples [24], ethanol in drinks [25], sulfite in beverages [26], and ascorbic acid in natural fruit juices [27].

In Vietnam, the coffee sector is estimated to provide a livelihood for approximately 2.6 million people, with the export value accounting for 3% of Vietnam's national GDP in 2014 [28]. Coffee has played a significant role in Vietnam's agriculture and economy. Since 2000, Vietnam has been known as the world's second-largest exporter and producer of coffee after Brazil [28]. As mentioned, the roasting degrees during the processing period perform certain effects on the phenolic compounds. For those bioactive components, there is not only the decomposition by high temperature, resulting in the decreased phenolic contents, but also the formation of other constituents with antioxidant capacity, which would potentially compensate for the chemical loss by applied roasting temperature. Such phenomenon was observed several publications [12,17,18]. Therefore, due to the different chemical processes that happened during the roasting period and TPC determination as a simple analytical method to reflex the bio-active components, it is necessary to monitor the TPC changes of the raw materials, the intermediate products during the processing, and the final products, especially for the coffee producers. Within the current context, there have still been certain limitations in the analytical abilities of non-expertized individuals, especially the lack of analytical chemistry background and the barrier to equipping with specific analytical instruments. As a common portable device, a smartphone performs the advantage of being applied in analytical approaches. Smartphone-based methods provide other tools for screening or monitoring the total phenolic contents in coffee during the processing as well as preliminary tests for final products before sending them to commercial accredited laboratories, which helps to save time and reduce production cost. Additionally, when the smartphone-based methods are fully developed, consumers can also use their smartphones to check their purchased items or to determine the TPCs in their coffee cups. In the present study, an inexpensive self-designed colorimetric box combined with an Android smartphone was designed to determine the TPCs in coffee using Folin-Ciocalteu as the complexing reagent. Several parameters related to photo capturing were investigated before evaluating the analytical method performance on the smartphone, in which the UV-Vis was employed as a standard method for comparison. The smartphone-based method was applied to specific Vietnamese coffee products to investigate their TPCs according to their varieties and roasting levels. To the best of our knowledge, the present study is among the first to use a smartphone to develop an analytical device for the determination of the TPCs in Vietnamese coffee products, especially, in the context that the publications on scientific data related to the internal compositions of Vietnamese coffee have still been limited.

2. Experimental

2.1. Sampling and total phenolic compound extraction

The roasted coffee beans were collected from a coffee producer in Lam Dong Province (Vietnam), transported to the laboratory, and endured the grinding process to achieve roasted ground coffee samples. There are a total of 12 separate samples belonging to two varieties of Arabica and Robusta. Three different roasting levels, i.e., light, dark, and medium, were applied for these products. Generally, during the roasting period, the green coffee beans were heated from 200 °C to 240 °C within 10–15 min, depending on the roasting levels, which were primarily evaluated by observing the color and appearance changes of the coffee beans by the producers [29]. For long-term storage, the coffee products were stored in a refrigerator at around –20 °C to minimize the changes in their internal composition. Before analysis, the roasted ground coffee products were kept in non-permeable bags sealed under a vacuum, stored at 25 °C, at 70% humidity, and avoided direct sunlight.

Ground coffee samples were extracted based on ISO 14,502–1 (2005) [30] with some modifications. Briefly, 0.2 (\pm 0.001) g of ground coffee sample was weighed into a 15-mL polypropylene (PP) centrifuge tube with a cap. Then, 10.00 mL of the extraction solvent consisting of 70% ν/ν methanol (MeOH, analytical grade, Merck, Germany) in deionized water (DIW, Millipore, USA) was added to conduct the mixture vortex for 1 min before the extraction in a water bath at 70 °C for 20 min. The tube was centrifuged at 4500 r min⁻¹, and the liquid was transferred into a 50-mL volumetric flask. The extraction procedure was repeated one more time, and the sample liquid was collected and combined into the volumetric flask before making up to the calibration mark by extraction solvent (70% ν/ν MeOH in DIW). The sample liquid was filtered through a 0.45- μ m PTFE membrane and diluted 20 times by DIW to proceed to the colorimetric assay and measurement for determining the total phenolic contents, expressed as mg GAE g⁻¹ on a dried weight basis or DW.

The colorimetric procedure was carried out through the chemical reaction between the phenolic compounds and the Folin-Ciocalteu reagent (Merck, Germany) as performed in ISO 14,502–1 (2005) [30] with some minor changes described in detail by Anh-Dao et al. (2021) [17]. However, the reaction time in the current study was aimed to reduce from 60 to 30 min to save the analysis time.

2.2. Self-designed colorimetric box

The reading of different color channels (RGB model) was performed by the freely available application of RGB color detector (version 3.0.7) from the Google Play Store. This application was installed on the Oppo A54 smartphone (Parameters: Android 10; CPU: Octa-core MediaTek Helio P35; display: 6.5 in., IPS LCD, HD+ (720 \times 1600 Pixels); main camera: 13 megapixels). A self-designed colorimetric box (12 \times 8 cm, length \times width \times height) was constructed from an paper box covered outside with a black plastic board. A cuvette or sample holder

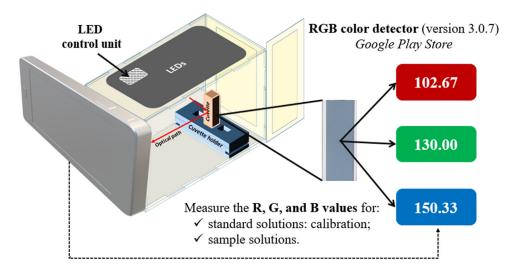


Fig. 1. Design of the colorimetric box.

was placed through the box top side to keep the 3.5-mL glass cuvette (Hellma, Germany, light path: 10 mm, outside dimension: 45 × 12.5 × 12.5 mm, inside width: 9.5 mm, base thickness: 1.5 mm). The observing window $(4 \times 2 \text{ cm})$ was designed at the opposite side of the cuvette to capture and analyze the images' color. The inside box is illuminated by light-emitting diodes (LEDs) on the inside top of the box, and there is a control unit to adjust the LEDs' intensities and turn the LEDs on/off (Fig. 1). The imaging area was fixed at the center of the sample/standard liquid column. The distance from the smartphone to the sample holder/cuvette and box inside brightness were investigated and kept constant throughout the experiments, i.e., 10 cm and 14,000 Lux (discussed later), respectively. The absolute values obtained from R, G, and B color channels were used as the measurement basis. The analytical response (calibration and sample analysis) was calculated as the color intensities of blank solution subtracted by those of each standard/sample solution (for each color channel) to demonstrate the increasing analytical response according to the rising concentrations as in the UV-Vis spectrophotometry.

2.3. Proposed analytical method performance

The present study evaluated and compared the analytical methods from three color channels with their application potentials. The analytical method performance was evaluated based on the guidelines and requirements of Appendix F of AOAC (2016) [31]. The calibration curves were obtained by plotting the milligrams of gallic acid mass equivalence at various concentrations, mg GAE L-1 (x) vs. their analytical responses (y) based on the linear relationship (y = axe + b). The limit of detection and quantification (LOD and LOQ) were calculated by the following equations: LOD = \bar{x} + 3SD and LOQ = 3LOD [32], in which \bar{x} and SD are, respectively, the estimated average concentration and the standard deviation values obtained from simultaneously analyzing 11 separate blank samples. The intra-day and inter-day precision were assessed through the calculation of the relative standard deviation for six replicates (n = 6) within one day (RSD_r) and three separate days (RSD_R). Spiked samples were used for recovery tests, in which the spiked concentrations for all samples were comparable to the estimated TPC in each sample. Gallic acid standard solution was used to spike to the ground coffee samples prior to the extraction process. Besides, to ensure the accuracy, t-test was applied to discover any statistical difference between the smartphone-based method and the standard UV-Vis [17,18], i.e., UV-Vis 1800 (Shimadzu, Japan) operated by UVProbe 2.34 software, the quantification wavelength of 765 nm, and a 10-mm light path glass cuvette as the measurement cell. There should be no significant difference between the two methods at the significance level P of 0.05 and

degree of freedom df = $n_1 + n_2 - 2 = 6 + 6 - 2 = 10$, in which there were six replicates for both the smartphone-based method ($n_1 = 6$) and UV–Vis ($n_2 = 6$). The critical value *t*-value (from the *t*-distribution table for the two-tailed test) is 2.23 for df = 10 and P = 0.05, and the calculation equations were obtained from the literature [32].

The analytical data were carried out in triplicates (n = 3), except for the statistically significant difference experiments (n = 6), and analyzed by Microsoft Office Excel (version 2205, Microsoft 365 Subscription). The sample results were expressed as mean value \pm standard deviation (SD). Relative standard deviations (RSDs) were calculated to evaluate and ensure the precision of measurement among runs.

3. Results and discussion

3.1. Distance from the smartphone camera to the sample holder

The effects of distance from the camera to the cuvette on the measured intensities for each color channel were investigated, in which the focal length of the smartphone camera was 26 mm. The basic parameters of the smartphone camera were applied, i.e., fixed-focused without flash, no image zoom, no photography effects, and single image mode. The results in Fig. 2 demonstrate that the changes in the capturing distance from 5 cm to 30 cm did not remarkably influence the measured intensities for all color channels. However, the precision was different for each distance, in which 10 cm exhibited the most favorable precision compared to other distances (see the error bars). This may be due to the blurred captured images for very close (5 cm) and longer distances (especially up to 30 cm). Moreover, longer focal lengths indicate fewer benefits because they require bigger sizes of the designed colorimetric boxes. Therefore, in the present study, 10-cm was applied as the distance from the smartphone camera to the cuvette, and the colorimetric box was designed to have 12-cm length to favor that distance. Other smartphones could be used with the designed colorimetric box; however, there might be certain variations in the obtained RGB values due to the differences in the camera operating parameters, e.g., resolution. Therefore, measuring the standard solutions to establish the calibration curve for each smartphone is necessary before analyzing the real sam-

3.2. Constant illumination inside the colorimetric box during color capturing

The brightness was measured by Lux Light Meter Software (installed on the smartphone) and expressed by Lux unit, in which the higher Lux values indicate brighter conditions. The constant light inside the closed

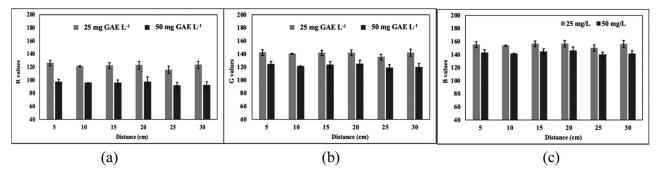


Fig. 2. Effects of focal lengths on the measured color intensities from (a) R, (b) G, and (c) B channels.

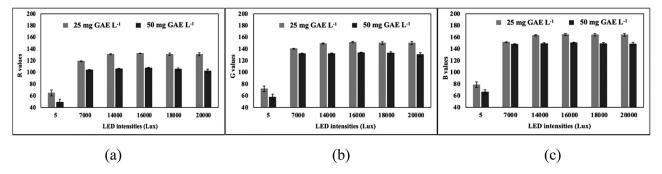


Fig. 3. Effects of brightness intensities on the measured color intensities from (a) R, (b) G, and (c) B channels.

colorimetric box during image capturing was reported to be necessary to ensure sensitivity and precision [26,33-35]. However, the brightness intensity has to be carefully investigated because it is different based on the box design and light sources used. In the present study, LEDs were used as the light source to maintain the constant illumination inside the colorimetric box, and six different LED intensity levels were evaluated. The closed box was designed to minimize the influences of the surrounding light, i.e., sunlight, light bulbs in the rooms, etc. The results in Fig. 3 demonstrate the brightness affected both the measured intensities and their precision for all channels. Proper LED intensities provide enough illumination for photo capturing and minimize the contribution of dark conditions in the box to the measured color intensities. This was supported by the very low recorded color intensities for 5 Lux (Fig. 3) and the increasing intensities for 7,000 Lux. Additionally, the level of 7,000 Lux might provide not enough (as mentioned) and not homogeneous light distribution inside the colorimetric box, leading to relatively lower measured color intensities than those obtained from the conditions of 14,000 Lux onwards. However, poorer precision was observed when the LED intensities were increased to 18,000 Lux and 20,000 Lux (see larger error bars, especially for 20,000 Lux), in which there might be excess brightness, resulting in data errors from the color intensity recording (noted that only one fixed point in the sample cuvette was used as the photo capturing target). This was supported by observing some shadows on the sample cuvette, particularly for higher concentrations. Therefore, to ensure constant and enough brightness inside the colorimetric box, to avoid undesired excess brightness, and to save the LED ages/energy, 14,000 Lux was used.

3.3. Reaction time reduction

The colorimetric assay was carried out based on the reaction between the phenolic compounds in the coffee extract and $10\% \,\nu/\nu$ Folin-Ciocalteu reagent to form the molybdenum blue and the tungsten blue [36]. It could be practically observed that the blue color of the formed complex appeared after mixing the reaction solution for more than 3 min, then the blue color would increase according to the rising reaction time. In the present study, the reaction time was investigated by

calculating the bias percentage (% bias) using the following equation:

% bias =
$$\frac{I_i - I_{60}}{I_{60}}$$
 (1)

whereas, I_i and I_{60} are the analytical response (measured RGB values) recorded after mixing the reaction solution for "i" minutes (i = 5, 10, 15, 20, 25, 30, 35, 40, 45, 50, and 60) and at the 60th min as the standard basis in ISO 14,502–1 (2005) [30].

The results from Fig. 4 indicate that the % bias values were lower than 3.0% from 25 min onwards for all three color channels and two investigated concentrations of 25.0 mg GAE $\rm L^{-1}$ and 50.0 mg GAE $\rm L^{-1}$. Therefore, 25–30 min could be chosen as the reaction time to save the analysis time, be more beneficial for *in situ* measurement, and help to improve the sample throughput. This was supported by several publications demonstrating 30 min as the proper reaction time for complex stability before further spectrophotometric measurement [36–38].

3.4. Analytical method characteristics from R, G, and B channels

In this current study, the absolute intensity values of three color channels (R, G, and B) were used to calculate the analytical response, then to propose three analytical approaches from the smartphone-based method for determining the TPCs in coffee products. The performance of these approaches was evaluated and compared with UV-Vis spectrophotometry as a standard method. The results in Table 1 indicate that the R channel demonstrated the highest sensitivity, i.e., the highest slope (a) value, since the color developed by the formed complex is from green to blue, and the complementary color is orange-red [39]. The sensitivity was followed by G and B channels, respectively. As expected, the linear ranges of the three-color channel, starting from 10.0 mg GAE L⁻¹ to the highest concentration that the relation between concentrations and corresponding analytical responses is still linear, performed in the descending order of B > G > R (Fig. S1). Therefore, for high concentrations of phenolic compounds in the extracts, the analytical method from the B channel could be considered more favorable for reducing the dilution factors. Moreover, the most extended linear range was suitable for wide ranges of TPCs in different coffee matrices. To achieve better sensitivity,

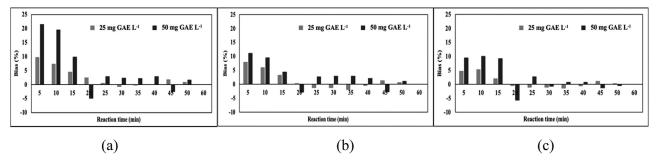


Fig. 4. Effects of reaction time on the formed complexes measured by (a) R, (b) G, and (c) B channels. Average values were calculated from the TPC results obtained from the UV–Vis and three color channels (smartphone-based method).

Table 1 Performance of analytical methods.

Criteria	UV-Vis 1800	Smartphone-based methods		
		R channel	G channel	B channel
Linear range (mg GAE L ⁻¹)	10.0–140.0	10.0-70.0	10.0-100.0	10.0-120.0
LOD (mg GAE L ⁻¹)	1.80	1.20	1.80	2.20
LOQ (mg GAE L ⁻¹)	5.40	3.60	5.40	6.60
Slope (a)	0.0117	1.6486	1.2234	0.7937
Intercept (b)	0.00	-1.1143	-2.1188	-1.6541
R^2	0.999	0.9987	0.9991	0.9979
Intra-day precision (%RSD _r)	1.84	2.00	2.01	2.02
Inter-day precision (%RSD _R)	1.95	2.10	2.15	2.16
Recoveries	98.9-101	99.1-101	99.3-101	99.3-101
Experimental t-value*		1.96	1.08	1.71
Critical <i>t</i> -value ($P = 95\%$)	2.23			

^{*} the highest value.

the R channel is preferable for other plants, fruit, and food types containing lower TPCs. Within the obtained linear ranges, the goodness of linearity was observed for all color channels, in which R^2 values were all higher than 0.995. Theoretically, the intercept (b) in the linear regression equation (y = axe + b) is expected to be low (in absolute values), especially when low analyte concentrations (around LOQ) were determined by the calibration curve. In the present study, the absolute values of the intercepts (b) were 1.1143, 1.6541, and 2.1188 for R, B, and G color channels, respectively (Table 1), which were low, compared to the analytical response (see Fig. S1). Moreover, the TPCs were generally ranged in the relatively high concentrations for coffee products. Therefore, the obtained regression equations from three color channels in the smartphone-based method could be reliably applied for TPC analysis.

The precision for both intra-day (%RSD_r) and inter-day (%RSD_R) was acceptable in all analytical methods, according to Appendix F of AOAC (2016) [31]. Spiked coffee samples and a t-test (P=95%) were used to evaluate the accuracy of the analytical methods obtained from three different color channels compared to the standard UV–Vis method. The results demonstrate that all spiked samples exhibited favorable recoveries (from 99.1% to 101% vs. 98.9%–101% for smartphone-based and UV–Vis, respectively). Additionally, with the critical t-value of 2.23, all proposed analytical methods from R, G, and B channels demonstrated no statistically significant difference, i.e., all experimental t-values were lower than 2.23 (significance level of 0.05) [32] (Table S1). Therefore, the smartphone-based analytical methods with their advantages, e.g., inexpensive, simple, portable, and easy operation, could be applied to estimate and determine the TPCs in various coffee products belonging to multiple roasting levels and origins.

3.5. Variability of total phenolic contents in coffee products

Fig. 5 illustrates the bar graph representation of the TPC determination in coffee products by the smartphone and the standard spec-

trophotometer. The figure demonstrates that the experimental results obtained from both analytical tools exhibit their similarity, and there were no statistically significant differences (mentioned earlier). Besides, the highest relative standard deviation (%RSD) obtained from the smartphone methods was only 0.81%, indicating the favorable repeatability compared to the standard spectrophotometric approach (%RSDs ranging from 0.12% to 2.0%) [31]. The average values of the three-color channel and UV-Vis were calculated for each sample to observe the variation in TPCs among different coffee products. The results in Fig. 5 demonstrate that, as a general trend, robusta coffee performed higher TPCs than those of arabica (35.1-46.0 mg GAE g^{-1} vs. 27.0-37.0 mg GAE g^{-1}). A similar observation was reported in several publications on TPCs in coffee products [17,18,40-43]. The higher TPCs in robusta coffee could be attributed to its higher concentrations of chlorogenic acid (CGA), which is very rich and specific in coffee, contributing to the results of TPCs [44,45]. Besides the higher condensed tannins, the main phenolic compounds in robusta coffee products could also account for their higher recorded TPCs [6]. Moreover, the roasting processes perform certain effects on the TPCs in coffee. Within the same coffee variety, the TPCs were the highest for light roasting levels, followed by medium and dark modes (robusta: 44.1-46.0 > 38.3-39.9 > 35.1-36.8 mg GAE g⁻¹ and arabica: 34.8-37.0 > 32.4-33.6 > 27.0-30.0 GAE g^{-1}). This might be explained because the prolonged roasting time at higher temperatures (medium and dark modes compared to light mode) led to the decomposition of heat-sensitive phenolic compounds. However, the gaps in TPCs among the three roasting levels were observed to be relatively narrow, possibly due to the Maillard reactions occurring during the roasting period. Such reactions produce other phenolic compounds, such as melanoidins and quinic acid [29,46-48]. Therefore, in the roasting process, the decomposition by heat and the formation of different phenolic compounds would happen simultaneously in two opposite directions. However, the formation side might not compensate enough for the loss by high temperature, which was supported by the lower TPCs according to the rising roasting levels in this current study.

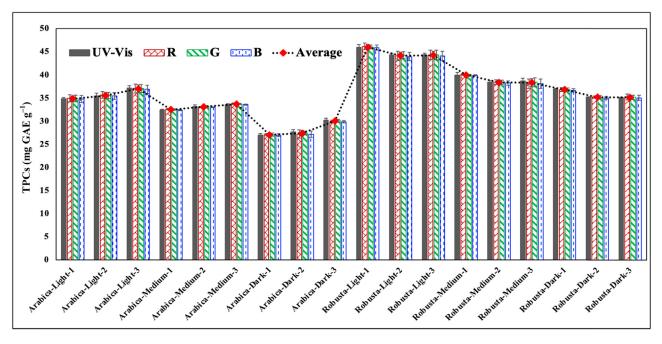


Fig. 5. Total phenolic contents of arabica and robusta coffee products regarding their different roasting levels.

4. Conclusions

The proposed analytical method based on a self-designed colorimetric box with a smartphone using RGB Color Detector App. demonstrates the favorable application for determining the total phenolic contents (TPCs) in coffee. The box was designed from an old tea box using the optimized parameters. The smartphone-based methods increase the performance of the TPC analysis in terms of their simplicity, portability, low cost, and easy accessibility to users due to the popularity of the smartphone. The quantification could be carried out using all threecolor channels with proper dilution factors for each condition (due to different sensitivities). However, for phenolic compound-rich sample matrices like coffee (and tea), the B channel should be used as the analytical response for quantification. Then, the availably set up method as a good alternative approach could serve as a powerful tool to estimate the phenolic contents immediately right in coffee (and tea) cups under different brewing conditions, especially for not adequately trained individuals in analytical chemistry, e.g., coffee producers and consumers.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRediT authorship contribution statement

Le-Thi ANH-DAO: Methodology, Validation, Writing – original draft, Investigation. Nguyen THANH-NHO: Writing – original draft. Bui HUU-TRUNG: Investigation, Software, Formal analysis. Nguyen TIEN-GIANG: Investigation, Software. Thach UT DONG: Investigation, Software. Nguyen QUOC-DUY: Investigation, Validation. Nguyen QUANG-HIEU: Investigation. Nguyen LE-VY: Investigation. Nguyen-Thi THANH-DIEU: Investigation. Dien Vu Thanh TO: Investigation. Do MINH-HUY: Software, Validation, Formal analysis, Writing – review & editing. Nguyen CONG-HAU: Conceptualization, Formal analysis, Writing – review & editing.

Acknowledgements

The authors would like to thank the Faculty of Environmental and Food Engineering, Nguyen Tat Thanh University, Ho Chi Minh City (Vietnam) for their assistance and support during this study.

Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.cjac.2023.100228.

References

- Samsonowicz M, Regulska E, Karpowicz D, Leśniewska B. Antioxidant properties of coffee substitutes rich in polyphenols and minerals. Food Chem 2019;278:101–9.
- [2] Dirks-Naylor AJ. The benefits of coffee on skeletal muscle. Life Sci 2015;143:182–6.
 [3] Brzezicha J, Błażejewicz D, Brzezińska J, Grembecka M. Green coffee VS dietary supplements: a comparative analysis of bioactive compounds and antioxidant activity. Food Chem Toxicol 2021;155:112377.
- [4] Kim Y, Je Y, Giovannucci E. Coffee consumption and all-cause and cause-specific mortality: a meta-analysis by potential modifiers. Eur J Epidemiol 2019;34:731–52.
- [5] Surya Prakash N, Combes MC, Somanna N, Lashermes P. AFLP analysis of introgression in coffee cultivars (*Coffea arabica* L.) derived from a natural interspecific hybrid. Euphytica 2002;124:265–71.
- [6] Hečimović I, Belščak-Cvitanović A, Horžić D, Komes D. Comparative study of polyphenols and caffeine in different coffee varieties affected by the degree of roasting, Food Chem 2011;129:991–1000.
- [7] Bottazzi D, Farina S, Milani M, Montorsi L. A numerical approach for the analysis of the coffee roasting process. J Food Eng 2012;112:243–52.
- [8] Fabbri A, Cevoli C, Alessandrini L, Romani S. Numerical modeling of heat and mass transfer during coffee roasting process. J Food Eng 2011;105:264–9.
- [9] Ruosi MR, Cordero C, Cagliero C, Rubiolo P, Bicchi C, Sgorbini B, Liberto E. A further tool to monitor the coffee roasting process: aroma composition and chemical indices. J Agric Food Chem 2012;60:11283–91.
- [10] Macheiner L, Schmidt A, Karpf F, Mayer HK. A novel UHPLC method for determining the degree of coffee roasting by analysis of furans. Food Chem 2021;341:128165.
- [11] Kwak HS, Ji S, Jeong Y. The effect of air flow in coffee roasting for antioxidant activity and total polyphenol content. Food Control 2017;71:210–16.
- [12] Bobková A, Hudáček M, Jakabová S, Belej L, Capcarová M, Čurlej J, Bobko M, Árvay J, Jakab I, Čapla J, Demianová A. The effect of roasting on the total polyphenols and antioxidant activity of coffee. J Environ Sci Health B 2020;55:495–500.
- [13] Belguidoum K, Amira-Guebailia H, Boulmokh Y, Houache O. HPLC coupled to UV–Vis detection for quantitative determination of phenolic compounds and caffeine in different brands of coffee in the Algerian market. J Taiwan Inst Chem Eng 2014:45:1314–20.

- [14] Nzekoue FK, Angeloni S, Navarini L, Angeloni C, Freschi M, Hrelia S, Vitali LA, Sagratini G, Vittori S, Caprioli G. Coffee silverskin extracts: quantification of 30 bioactive compounds by a new HPLC-MS/MS method and evaluation of their antioxidant and antibacterial activities. Food Res Int. 2020;133:109128.
- [15] Mehari B, Chandravanshi BS, Redi-Abshiro M, Combrinck S, McCrindle R, Atlabachew M. Polyphenol contents of green coffee beans from different regions of Ethiopia. Int J Food Prop. 2021;24:17–27.
- [16] Fukushima Y, Tashiro T, Kumagai A, Ohyanagi H, Horiuchi T, Takizawa K, Sugihara N, Kishimoto Y, Taguchi C, Tani M, Kondo K. Coffee and beverages are the major contributors to polyphenol consumption from food and beverages in Japanese middle-aged women. J Nutr Sci 2014;3:e48.
- [17] Anh-Dao LT, Nhon-Duc L, Cong-Hau N, Thanh-Nho N. Variability of total polyphenol contents in ground coffee products and their antioxidant capacities through different reaction mechanisms. Biointerface Res Appl Chem 2021;12:4857–70.
- [18] Thanh-Nho N, Anh-Dao LT, Chi NTK, Cong-Hau N. Molecular absorption spectrophotometric method for determining total polyphenol contents in roasted ground coffee products. TNU J Sci Technol. 2021;226:124–32.
- [19] Mahato K, Chandra P. Paper-based miniaturized immunosensor for naked eye ALP detection based on digital image colorimetry integrated with smartphone. Biosens Bioelectron, 2019;128:9–16
- [20] Minh-Huy D, Anh-Dao LT, Thanh-Nho N, Nhon-Duc L, Cong-Hau N. Smart-phone-based digital images as a low-cost and simple colorimetric approach for the assessment of total phenolic contents in several specific Vietnamese dried tea products and their liquors. Food Chem 2023;401:134147.
- [21] Ledesma CM, Krepsky LM, Borges EM. Using a flatbed scanner and automated digital image analysis to determine the total phenolic content in beer. J Chem Educ 2019:96:2315–21.
- [22] Al-Nidawi M, Alshana U. Reversed-phase switchable-hydrophilicity solvent liquid-liquid microextraction of copper prior to its determination by smartphone digital image colorimetry. J Food Compos Anal 2021;104:104140.
- [23] Bhagat B, Jadeja V, Sharma P, Joshi K, Mukherjee K. Case study on the use of image analysis for the simple and inexpensive colorimetric detection of Fe(III) in water. Anal Methods 2020;12:4509–16.
- [24] Jing X, Wang H, Huang X, Chen Z, Zhu J, Wang X. Digital image colorimetry detection of carbaryl in food samples based on liquid phase microextraction coupled with a microfluidic thread-based analytical device. Food Chem 2021;337:127971.
- [25] Benedetti LPDS, dos Santos VB, Silva TA, Filho EB, Martins VL, Fatibello-Filho O. A digital image-based method employing a spot-test for quantification of ethanol in drinks. Anal Methods 2015;7:4138–44.
- [26] Pires dos Santos Benedetti L, Bezerra dos Santos V, Silva TA, Benedetti-Filho E, Martins VL, Fatibello-Filho O. A digital image analysis method for quantification of sulfite in beverages. Anal Methods 2015;7:7568–73.
- [27] Porto ISA, Santos Neto JH, dos Santos LO, Gomes AA, Ferreira SLC. Determination of ascorbic acid in natural fruit juices using digital image colorimetry. Microchem J 2019:149:104031.
- [28] Nguyen GNT, Sarker T. Sustainable coffee supply chain management: a case study in Buon Me Thuot City, Daklak, Vietnam. Int J Corp Soc Responsib 2018;3:1.
- [29] Asfaw G, Tefera M. Total polyphenol content of green, roasted and cooked Harar and Yirgacheffee Coffee, Ethiopia. J Appl Sci Environ Manag 2020;24:187–92.
- [30] ISO 14502-1 Determination of substances characteristic of green and black tea-Part 1: content of total polyphenols in tea-Colorimetric method using Folin-Ciocalteu reagent. 1st ed. Switzerland: International Organization for Standardization; 2005.

- [31] Appendix F guidelines for standard method performance requirements, association of official analytical chemists 2016. https://www.aoac.org/resources/guidelines-for-standard-method-performance-requirements/.
- [32] Ellison SL, Barwick VJ, Farrant TJD. Practical statistics for the analytical scientist: a bench guide. 2nd ed. Royal Society of Chemistry; 2009.
- [33] Didpinrum P, Siriangkhawut W, Ponhong K, Chantiratikul P, Grudpan K. A newly designed sticker-plastic sheet platform and smartphone-based digital imaging for protein assay in food samples with downscaling Kjeldahl digestion. RSC Adv 2021;11:36494–501.
- [34] dos Santos VB, da Silva EKN, de Oliveira LMA, Suarez WT. Low cost in situ digital image method, based on spot testing and smartphone images, for determination of ascorbic acid in Brazilian Amazon native and exotic fruits. Food Chem 2019;285:340-6.
- [35] Moonrungsee N, Pencharee S, Jakmunee J. Colorimetric analyzer based on mobile phone camera for determination of available phosphorus in soil. Talanta 2015;136:204-9.
- [36] Agbor GA, Vinson JA, Donnelly PE. Folin-Ciocalteau reagent for polyphenolic assay. IJFS 2014:3:147–56.
- [37] Blainski A, Lopes GC, De Mello JCP. Application and analysis of the Folin-Ciocalteu method for the determination of the total phenolic content from Limonium brasiliense L. Molecules 2013;18:6852–65.
- [38] Kamboj A, Gupta R, Rana A, Kaur R. Application and analysis of the Folin-Ciocalteu method for the determination of the total phenolic content from extracts of Terminalia bellerica. Eur J Biomed Pharm Sci 2015;2:201–15.
- [39] Franco MdOK, Suarez WT, dos Santos VB, Resque IS. A novel digital image method for determination of reducing sugars in aged and non-aged cachaças employing a smartphone. Food Chem 2021;338:127800.
- [40] Olechno E, Puścion-Jakubik A. Impact of brewing methods on total phenolic content (TPC) in various types of coffee. Molecules 2020;25:5274.
- [41] Perdani C, Pranowo D. Total phenols content of green coffee (Coffea arabica and Coffea canephora) in East Java. IOP Conf Ser Earth Environ Sci 2019;230:012093.
- [42] Trandafir I, Nour V, Ionica ME. Antioxidant capacity, phenolic acids and caffeine contents of some commercial coffees available on the Romanian market. Arch Latinoam Nutr 2013;63:87.
- [43] Sacchetti G, Di Mattia C, Pittia P, Mastrocola D. Effect of roasting degree, equivalent thermal effect and coffee type on the radical scavenging activity of coffee brews and their phenolic fraction. J Food Eng 2009;90:74–80.
- [44] Narita Y, Inouye K. Inhibitory effects of chlorogenic acids from green coffee beans and cinnamate derivatives on the activity of porcine pancreas α -amylase isozyme I. Food Chem 2011;127:1532–9.
- [45] Craig AP, Fields C, Liang N, Kitts D, Erickson A. Performance review of a fast HPLC-UV method for the quantification of chlorogenic acids in green coffee bean extracts. Talanta 2016;154:481–5.
- [46] Liu X, Xia B, Hu LT, Ni ZJ, Thakur K, Wei ZJ. Maillard conjugates and their potential in food and nutritional industries: a review. Food Front 2020;1:382–97.
- [47] Liu Y, Kitts DD. Confirmation that the Maillard reaction is the principle contributor to the antioxidant capacity of coffee brews. Food Res Int 2011;44:2418–24.
- [48] Opitz SE, Smrke S, Goodman BA, Keller M, Schenker S, Yeretzian C. Antioxidant generation during coffee roasting: a comparison and interpretation from three complementary assays. Foods 2014;3:586–604.