Revista Brasileira de Farmacognosia 29 (2019) 434–440

Brazilian Journal of Pharmacognosy
REVISTA BRASILEIRA DE FARMACOGNOSIA
www.elsevier.com/locate/bjp

Original Article

Convection combined microwave drying affect quality of volatile oil compositions and quantity of curcuminoids of turmeric raw material

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ARTICLE INFO

Article history:
Received 11 September 2018
Accepted 30 April 2019
Available online 22 June 2019

Keywords:
Convection drying
Curcuminoids
Microwave drying
Response surface methodology
Volatile oil

ABSTRACT

The aim of the work was to estimate the quality of some compositions in turmeric volatile oil and quantity of individual and total curcuminoids in turmeric powder dried under different conditions. Effect of convection time (0–30 min) and microwave time (20–60 min) on quality of volatile oil compositions and quantity of curcuminoids were investigated using computer software. Quality of volatile oil obtained from the hydrodistillation of dried turmeric was analyzed by gas chromatography–mass spectrometry. The quantity of individual curcuminoids (bisdemethoxycurcumin, demethoxycurcumin, and curcumin) and total curcuminoids were analyzed by high performance liquid chromatography. Ten volatile compounds in turmeric volatile oil were used to estimate the variation of their quality. Results showed that ar-turmerone, turmerone, and curlone were the three major compounds found in turmeric volatile oil. The quality of the ten volatile compounds varied depending on convection time and microwave time. The three principal curcuminoids were found in turmeric dried at long convection time and medium microwave time. However, curcumin was also found in high amount in turmeric dried at short convection time and long microwave time. Total curcuminoids in dried turmeric were equal to or greater than 5% (w/w) as stated in the Thai Herbal Pharmacopoeia. It was found that almost all of the drying procedure achieved the standard of the Thai Herbal Pharmacopoeia except at short convection time and short microwave time. In summary, convection combined with microwave drying affected the quality of volatile oil compositions and quantity of curcuminoids of turmeric raw material.

Introduction

Turmeric (*Curcuma longa* L., Zingiberaceae) is the herbal medicinal plant most used in Thailand for treatment of flatulence due to its high content of volatile oil. Furthermore, its extract is marketed as a modern medicine for the treatment of knee osteoarthritis due to its high content of curcuminoids. It can decrease inflammation and oxidative stress biomarkers in osteoarthritis patients (Srivastava et al., 2016). The efficacy and safety of turmeric extract are comparable to ibuprofen with fewer side effects (Kuptniratsaikul et al., 2009, 2014). Turmeric extract combined with diclofenac appeared to be more effective than diclofenac alone in the treatment of knee osteoarthritis, but was not statistically significant (Pinsornsak and Niempoog, 2012). Turmeric extract is able to suppress the secretion of cyclooxygenase-2 by synovial fluid monocytes of osteoarthritis patients similar to diclofenac sodium (Kertia et al., 2012). Not only curcuminoids, but the pure curcumin possesses anti-inflammatory activity, so it can be used for the treatment of rheumatoid arthritis. Curcumin alone has superior efficacy compared to diclofenac sodium alone or their combination (Chandran and Goel, 2012).

Drying is an important step to preserve the post-harvest herbal medicinal plant. Drying of plant raw materials requires attention to several parameters to maintain the quality of plant raw materials. Drying temperature influences the content of some phenolic compounds and carotenoids of *Chenopodium quinoa*. Heat applied to plant material may inactivate enzyme activity of the plant, so decomposition of phytochemicals is prevented. It can soften plant tissue, thus bioaccessibility or amount of compounds released from plant matrix is increased (Multari et al., 2018). Recently, microwaving has been used to dry plant raw materials in order to shorten the drying time compared to conventional air drying (Schmidt, 2018). However, the effect of the drying process on quality and quantity of chemical constituents must be evaluated to ensure the quality of plant raw materials. The aim of this work was to evaluate the effect of convection combined with microwave drying on the quality of volatile oil and quantity of curcuminoids of turmeric raw

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https://doi.org/10.1016/j.bjp.2019.04.006

material. The ten volatile compounds mostly found in volatile oil were selected to evaluate its quality under different drying conditions. Furthermore, three curcuminoids; bisdemethoxycurcumin, demethoxycurcumin, and curcumin were quantified to identify the best drying condition provided the best quality of turmeric raw material.

Materials and methods

Materials

Bisdemethoxycurcumin (BDMC), demethoxycurcumin (DMC), and curcumin (CUR) with purity 99.38%, 99.21%, 98.74%, respectively, were purchased from Chengdu Biopurify Phytochemicals Ltd., China. All solvents were HPLC grade and purchased from Honeywell-Burdick & Jackson, USA.

Plant sample

Turmeric rhizomes (*Curcuma longa* L., Zingiberaceae) used in this work originated from Thap Put District, Phangnga Province, Thailand. They were collected for 10 kg in March 2018. Plant samples were authenticated by Ajarn Nirun Vipunngeun, a plant taxonomist and lecturer at Department of Pharmacognosy, College of Pharmacy, Rangsit University. A voucher specimen was coded as CM-CL001-1-03-2018. They were placed at Drug and Herbal Product Research and Development Center, College of Pharmacy, Rangsit University.

Drying of turmeric rhizome

Turmeric rhizomes were cleaned, boiled for 30 min, and sliced. Sliced turmeric (500 g) with thickness of approximately 0.2–0.3 cm was dried by convection using hot air oven (RXH14-B, Changzhou Wangqun Pharmaceutical Machine Co., Ltd., China) at 60 °C, followed by microwaving using the microwave oven (EMS3288X, Electrolux, China) at level 7 (630 W). The microwave oven was modified to improve drying efficiency by connecting it with a roller chamber containing pore and three exhaust fans. The slices were dried with a specific time as shown in Fig. 1. The dried turmeric was ground, passed through a 40-mesh sieve, and stored in a dry place without excessive heat and excessive moisture.

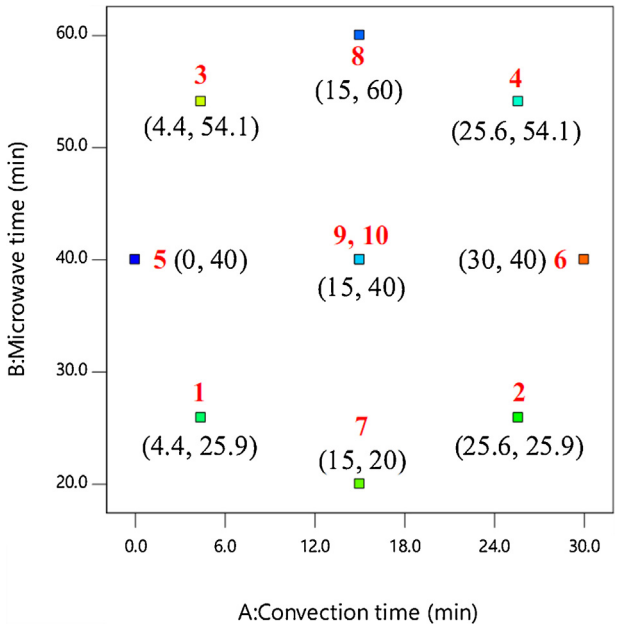

Fig. 1. Two-factor spherical composite experimental design.

Qualitative analysis of volatile oil composition

The volatile oil of turmeric powder was extracted using hydrodistillation technique. Turmeric powder (10 g) was added to water (100 ml) and distilled for 5 h using the Clevenger apparatus. Then, the receiving tube was allowed to cool to ambient temperature. The volatile oil was collected and stored in the refrigerator.

The gas chromatography–mass spectrometry (GC–MS) analysis was performed on GC-MS 7890A 5975C MSD (Agilent Technologies, USA). Mega-5MS column (30 m × 0.25 mm, i.d., 0.25 μm) (Mega S.r.l., Italy) was used for the separation. The sample was diluted with ethanol at a ratio of 1:20. The front inlet was kept at split mode (split ratio 1:10). The temperatures of the injector, auxiliary heater, MS source, and MS quadrupole, were 200, 250, 230, and 150 °C, respectively. The injection volume was 1 μl. Oven temperature was increased from 60 to 250 °C at 2.5 °C/min and held for 5 min. Mass spectra were scanned at 40–900 amu. The possible volatile oil compositions were based on the match with standard mass spectrum obtainable in the NIST 2011 GC-MS library database. Peak area and percent area of the peak were collected. The percent area of the peak was calculated by comparing peak area of interested peak to the summation of peak area of all peaks.

Quantitative analysis of individual and total curcuminoids

Stock solutions of standard BDMC, DMC, and CUR with a concentration of 1000 μg/ml were individually prepared using methanol as a solvent. Then, the mixed standards in five concentrations (*i.e.* 10, 25, 50, 75, and 100 μg/ml) were prepared. They were filtered and analyzed by high performance liquid chromatography (HPLC). Calibration curves of the three standards were constructed.

Turmeric powder (10 mg) was added to the 10-ml volumetric flask. Methanol was added and adjusted to the volume. They were sonicated for 30 min. The obtained solution was filtered and analyzed for the individual curcuminoids by HPLC. The total curcuminoids content was obtained from the summation of the three curcuminoids.

The HPLC analysis was performed on Agilent 1260 infinity (Agilent Technologies, USA). The separation was done using ACE Generix column (150 mm × 4.6 mm, i.d., 5 μm) (Advanced Chromatography Technologies Ltd., Scotland) with isocratic elution system containing acetonitrile and 1% acetic acid aqueous solution (55:45, v/v) at flow rate 1 ml/min. The column temperature was controlled at 30 °C. The injection volume was set at 10 μl and the injection was done by autosampler with needle washing. The signal was detected by a photodiode array detector at 425 nm.

Optimization procedure

The spherical composite experimental design was used for the optimization. Two factors were investigated *i.e.* convection time (X_1) and microwave time (X_2). Peak area of ten major peaks of volatile oil was monitored and used to construct the 3D response surfaces using Design-Expert® software version 11 (Stat-Ease, Inc., USA). The other responses (BDMC content (Y_1), DMC content (Y_2), CUR content (Y_3), and total curcuminoids content (Y_4)) were monitored. The 3D response surfaces were constructed. The predicted and actual values were plotted and the coefficient of determination (R^2) was reported. The internally studentized residual and run number were also plotted. The optimal condition was selected based on desirability function (Bezerra et al., 2008). Finally, overlay plot was constructed to display the condition that total curcuminoids content equal to or higher than 5% (w/w) as stated in

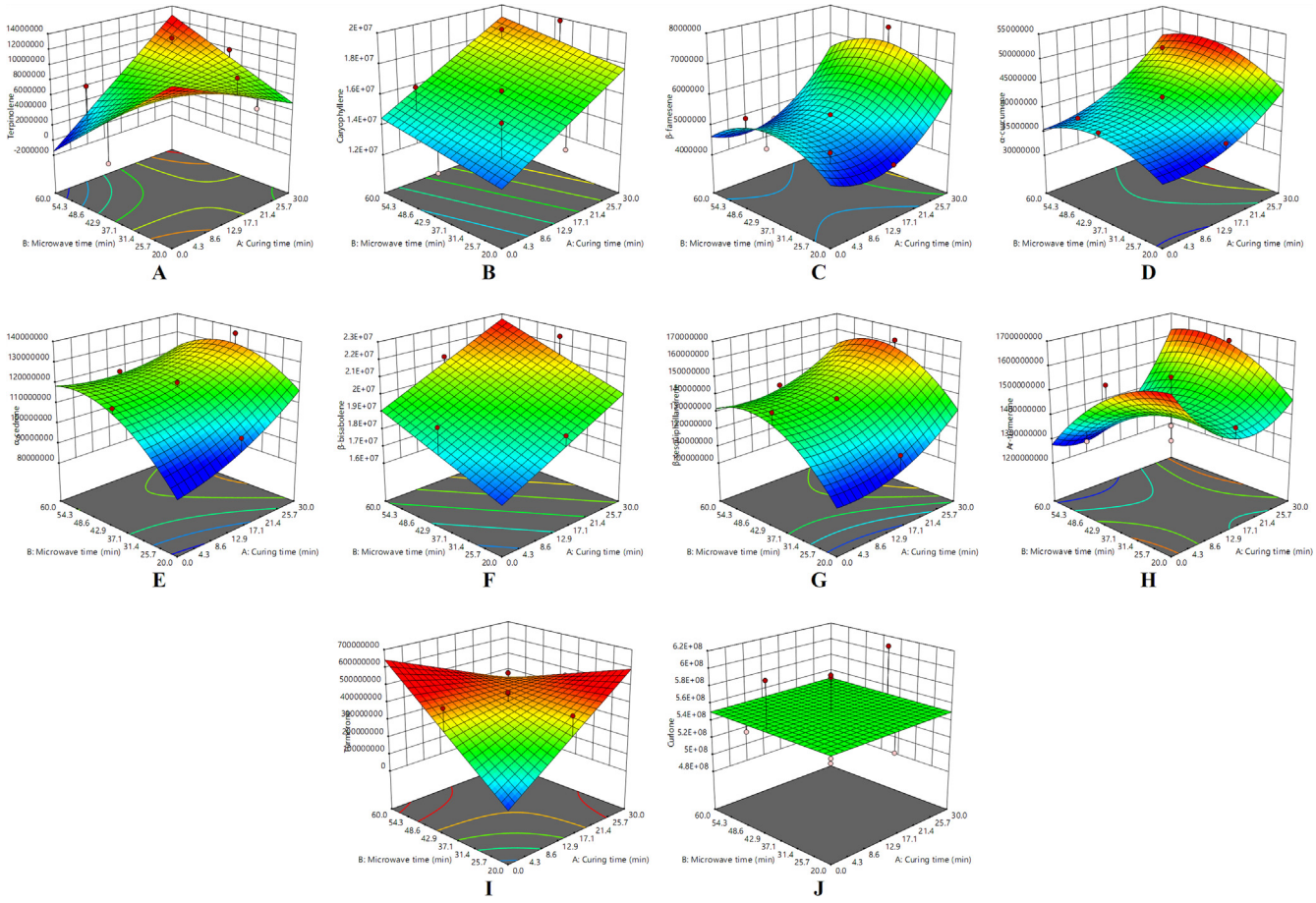

Fig. 2. 3D response surfaces of the model conditions of (A) terpinolene, (B) caryophyllene, (C) β-farnesene, (D) α-curcumene, (E) α-cedrene, (F) β-bisabolene, (G) β-sesquiphellandrene, (H) ar-turmerone, (I) turmerone, and (J) curlone.

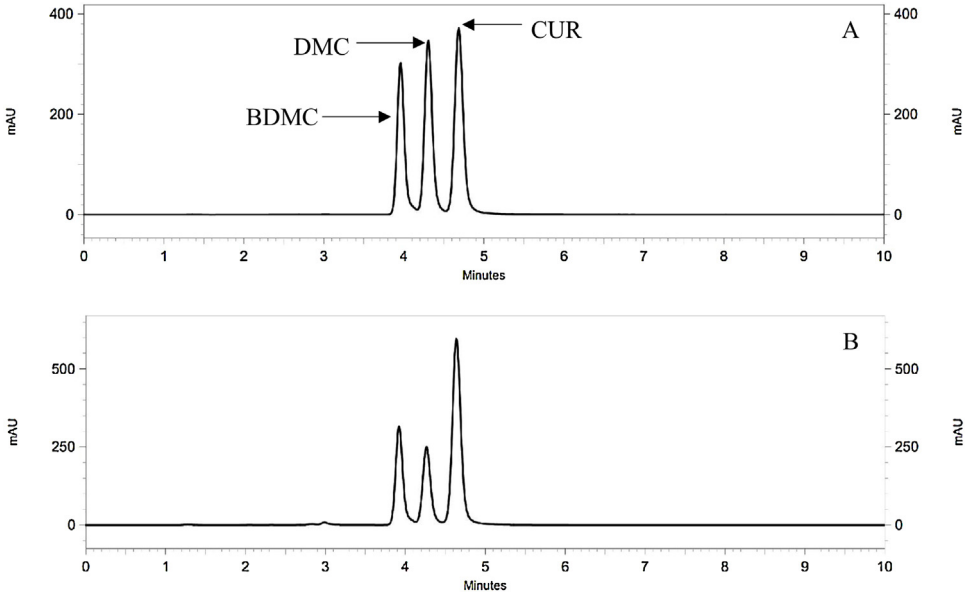

Fig. 3. HPLC chromatograms of (A) mixed standard curcuminoids (25 μg/ml) and (B) turmeric extract of Condition 9.

the Thai Herbal Pharmacopoeia (Department of Medical Sciences, 2017).

Results and discussion

Quality of some constituents in turmeric volatile oil and quantity of individual and total curcuminoids in turmeric powder dried under different conditions were determined in this work. According to the percent area from GC–MS determination, principal compounds in volatile oil of turmeric were ar-turmerone (43–49%), turmerone (13–16%), and curlone (17–18%), respectively. The results are comparable to the report of Leela et al. (2002): ar-turmerone, curlone, and turmerone were 31.1, 10.6, and 10%, respectively. Thongphasuk and Thongphasuk (2013) reported that ar-turmerone (32%), α-turmerone (16%), and β-turmerone (13%) were major volatile compounds. Some differences were found in turmeric volatile oil from Iran; it contained ar-turmerone (68.9%), α-turmerone (20.9%), and α-phellandrene (2.2%) (Asghari et al., 2009).

The peak area was used to estimate the content of individual volatile compound since it decreased the effect of other constituents compared to the percent area method. Fig. 2 shows 3D response surfaces of the model conditions of the peak area of ten compounds found in volatile oil (*i.e.* terpinolene, caryophyllene, β-farnesene, α-curcumene, α-cedrene, β-bisabolene, β-sesquiphellandrene, ar-turmerone, turmerone, and curlone). The content of volatile oil constituents was affected by the drying condition. Different convection time and microwave time revealed different content of individual volatile compounds except for curlone. However, the exact content of the volatile oil constituents was not determined because of the qualitative nature of the analysis. Thus, the condition for drying turmeric should be the same to maintain the quality of the chemical composition of the volatile oil. The effect of microwave drying on volatile substances of herbal plants was previously reported. Di Cesare et al. (2003) reported that some microwave conditions could preserve some volatile substances (*i.e.* eucalyptol, eugenol, methyl eugenol, and linalool) better than traditional air-drying at 50 °C. But the method is less effective at preserving the volatile substances than is freeze-drying without blanching technique. In the case of garlic, Rao et al. (2007) reported that microwave drying reduced drying time from 8 h by tray drying to 0.25 h by microwave drying. Microwave drying increased the concentration of diallyl disulfide and diallyl tetrasulfide, but decreased the concentration of diallyl trisulfide and allyl methyl trisulfide, relative to fresh garlic. Moreover, microwave drying provided similar quality of garlic compared to cabinet tray drying. Kubra and Rao (2012) reported that the volatile oil content of ginger dried by microwave was comparable to convection drying – approximately 3% (v/w). Moreover, the principal volatile compound, zingiberene, was increased relative to the fresh sample.

Fig. 3 shows the HPLC chromatograms of mixed standard curcuminoids and turmeric extract. BDMC eluted first followed by DMC and CUR, respectively. Turmeric extract contained greater amount of CUR than BDMC and DMC, respectively. Turmeric powder contained 2.3–3.6% CUR, 1.5–2.3% BDMC, and 1–1.6% DMC. Total curcuminoids content was 4.8–7.3% (w/w). The slightly different results were found in previous reports. CUR was mostly

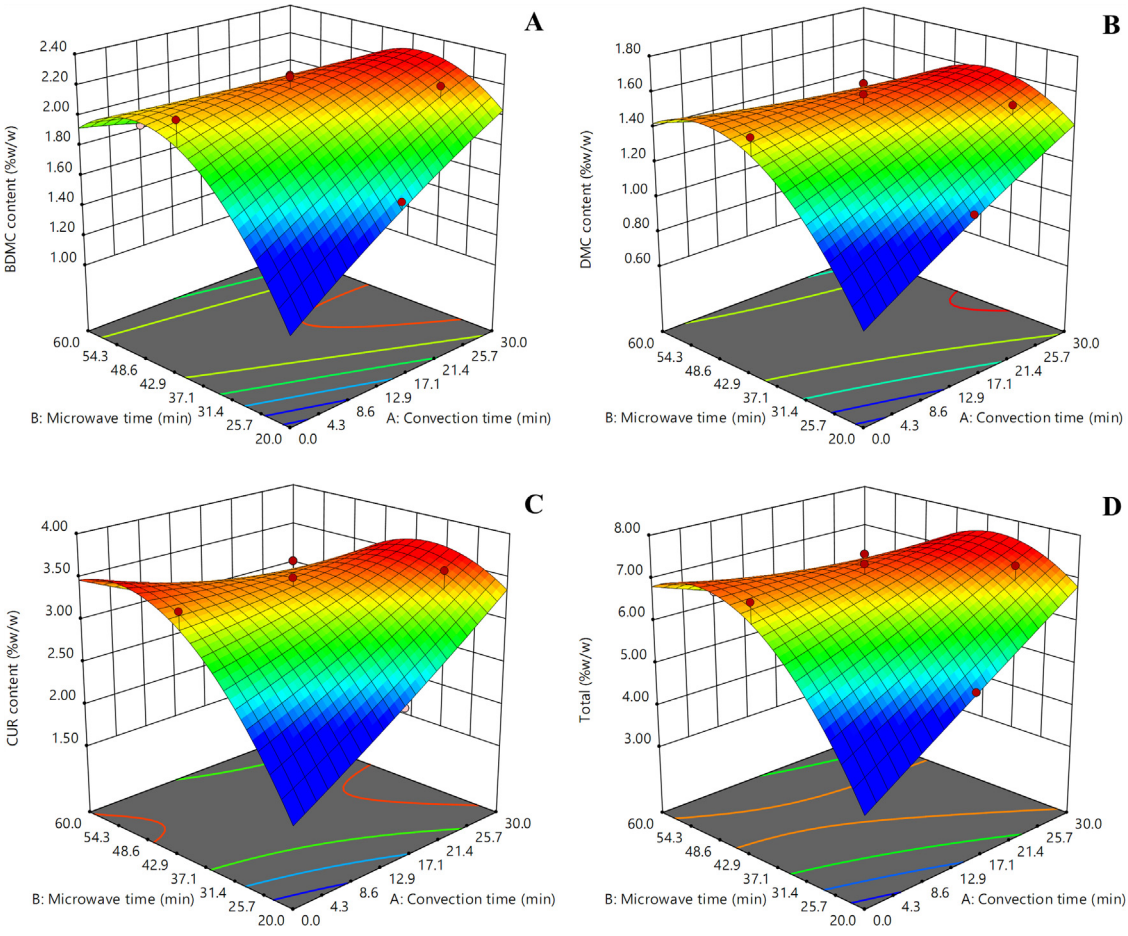

Fig. 4. Response surfaces of the model conditions of (A) BDMC, (B) DMC, (C) CUR, and (D) total curcuminoids content.

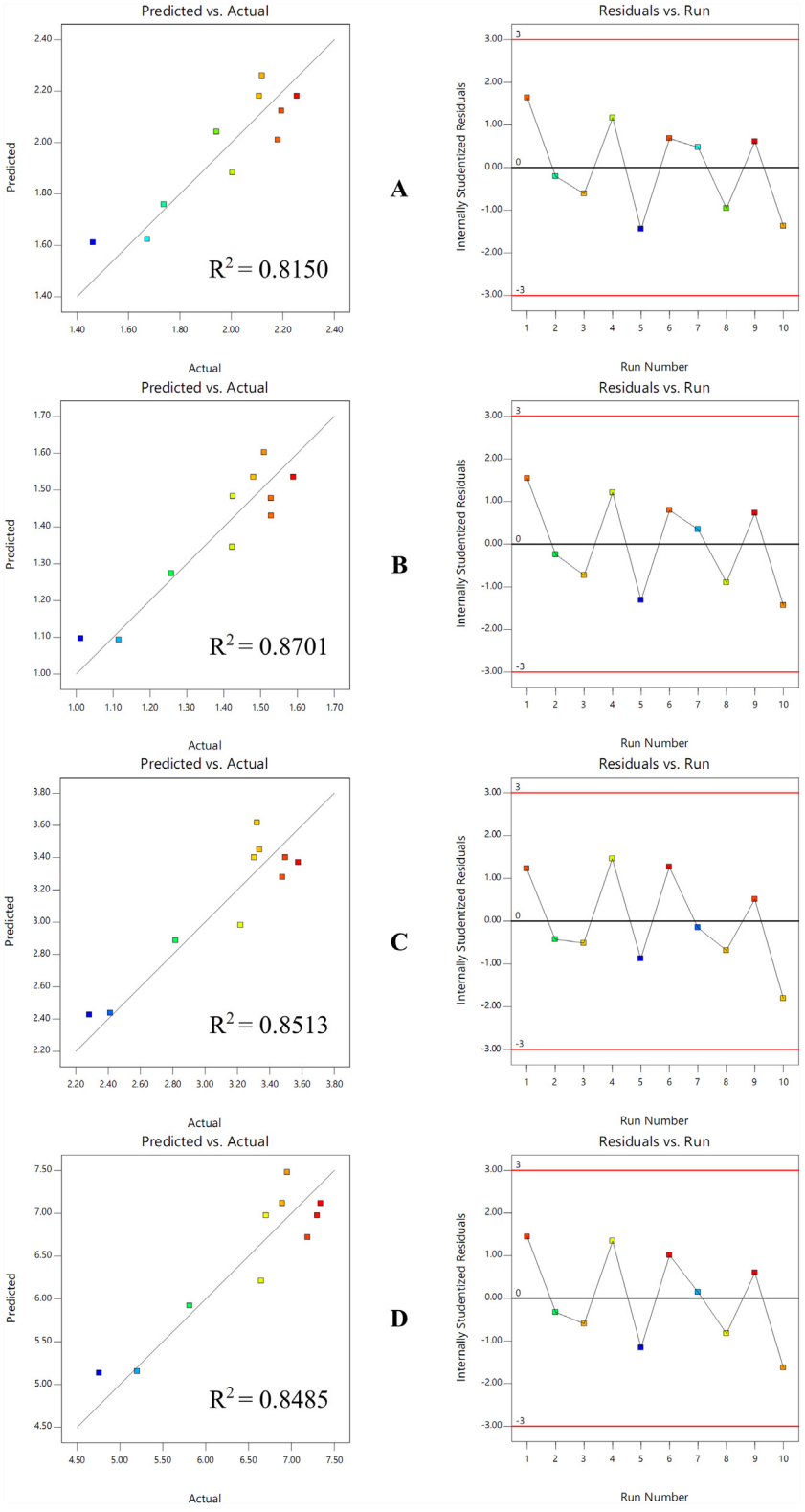

Fig. 5. The predicted vs actual value plots (left) and internally studentized residual vs run number plots (right) of the model condition of (A) BDMC, (B) DMC, (C) CUR, and (D) total curcuminoids content.

found follow by DMC and BDMC. Turmeric contained 1–5.7% CUR, 0.8–3.4% DMC, and 0.4–2.2% BDMC or 2.3–9.2% total curcuminoids were reported (Jayaprakasha et al., 2002). Ali et al. (2014) reported that turmeric powder contained 2.1% CUR, 0.5% DMC, and 0.1% BDMC. This order of content was also found in the report of Osorio-Tobón et al. (2016). They reported total curcuminoids of turmeric rhizome of 18.2–23.3 mg/g or 1.8–2.3% (w/w). CUR, DMC, and BDMC content were 10.2%, 5.9–7%, and 2.1–6.1%, respectively. The high content of total curcuminoids; 12–14.4% was previously reported

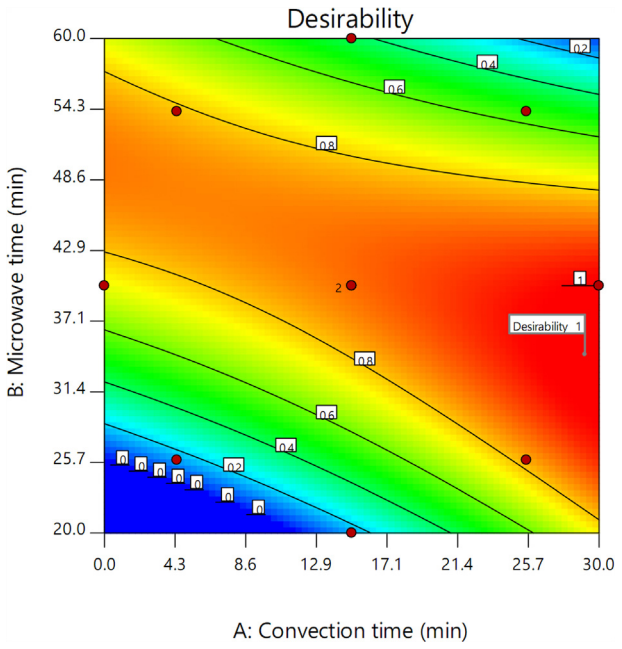

Fig. 6. Contour plot of desirability.

by our colleague (Monton et al., 2016). Fig. 4 shows the 3D response surfaces of the model conditions of BDMC, DMC, CUR, and total curcuminoids content. The three principal curcuminoids were found in greatest amount when the turmeric was dried at long convection time and medium microwave time. However, curcumin was found in greatest amount in turmeric dried at short convection time and long microwave time. Hirun et al. (2014) mentioned that a moderate temperature is important to preserve curcuminoids and other compounds. They reported that longer microwave drying time increased antioxidant activity due to the high content of curcuminoids. They also suggested that microwave-vacuum drying could inhibit enzymatic browning and improve physical appearance as well as maintain bioactive compounds. Furthermore, the optimal condition provided a good quality turmeric was a high microwave power (3500–4000 W) and long duration (27–30 min). Gagare et al. (2015) optimized the process parameters for curing and microwave drying of turmeric rhizomes. Turmeric rhizomes were boiling in 0.1% sodium carbonate solution for 15–45 min followed by microwave drying at power of 1000–2000 W. Curcumin content was decreased when microwave power was increased from 1000 W to 2000 W. The best quality turmeric (color uniformity, appearance, skin removal, and maintenance of curcumin content) was achieved with curing time of 30 min at 1500 W.

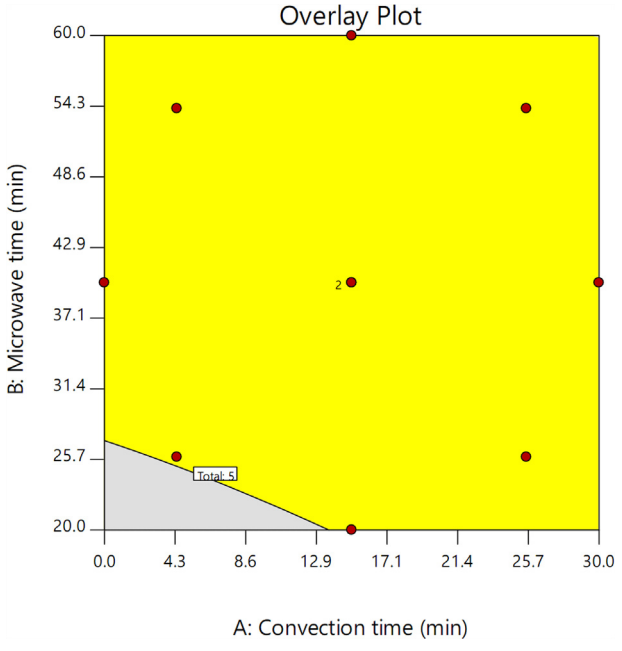

Fig. 7. Overlay plot, where the yellow area was the area that total curcuminoids content not less than 5.0% (w/w).

The relatively high R^2 between the predicted and actual value of model conditions of the three individual curcuminoid and total curcuminoids content was observed in this work (Fig. 5). The internally studentized residuals vs run number plots showed that the distribution of the data was within the 95% confidence interval; all data were distributed within the red line (Fig. 5). These results confirmed the reliability and stability of the computer software-based estimation (Duangjit et al., 2012, 2014). The actual equations used for estimation of each response: BDMC content (Y_1), DMC content (Y_2), CUR content (Y_3), and total curcuminoids content (Y_4) are shown below:

$$Y_1 = -0.7559 + 0.0592X_1 + 0.1181X_2 - 0.001X_1X_2 - 0.0002{X_1}^2 - 0.0012{X_2}^2$$

$$Y_2 = -0.6772 + 0.0430X_1 + 0.0878X_2 - 0.0009X_1X_2 - 0.0001{X_1}^2 - 0.0009{X_2}^2$$

$$Y_3 = -1.5393 + 0.0993X_1 + 0.1943X_2 - 0.0024X_1X_2 - 0.0002{X_1}^2 - 0.0018{X_2}^2$$

$$Y_4 = -2.9725 + 0.2015X_1 + 0.4003X_2 - 0.0043X_1X_2 - 0.0001{X_1}^2 - 0.0039{X_2}^2$$

The optimal condition was selected based on desirability function. The desirability value of 1 is indicative of the most desirable outcome (Bezerra et al., 2008). The best response was achieved when the high content of total curcuminoids was achieved. Turmeric dried at long convection time and medium microwave time provided the highest total curcuminoids content. Conversely, the desirability value equal to 0 (a completely undesirable response) was occurred at short convection and short microwave time (Fig. 6). The optimal condition that provided the greatest content of total curcuminoids was convection time and microwave time of 28.8 min and 37.4 min, respectively. This condition gave 7.5% (w/w) total curcuminoids content with desirability value of 1. The yellow area of the overlay plot in Fig. 7 represents the experimental region in which total curcuminoids content was equal to or higher than 5% (w/w), which meet the standard of the Thai Herbal Pharmacopoeia. Conversely, conditions of short convection time and microwave time (gray area) failed to meet the standard.

Conclusions

Convection combined microwave drying affected the quality of volatile oil compositions and quantity of curcuminoids of turmeric raw material. The BDMC, DMC, and CUR were found in greatest amount in turmeric dried at long convection time and medium microwave time. However, curcumin was also found in

large amount in turmeric dried at short convection time and long microwave time. The three individual curcuminoids that were assayed also represented the principal quantity of curcuminoid content under condition of long convection time and medium microwave time. Total curcuminoids content of dried turmeric raw material reached the standard of the Thai Herbal Pharmacopoeia when it was equal to or higher than 5% (w/w). Hence, total curcuminoids content failed the standard of the Thai Herbal Pharmacopoeia when turmeric was dried at short convection time and short microwave time.

Authors' contributions

CM as a project leader, designed the experiments, contributed to the experimental part, analyzed and interpreted the data, and drafted the manuscript. CL and LC analyzed and interpreted the data. All authors have read and approved the final version of the manuscript.

Conflicts of interest

The authors declare no conflicts of interest.

Acknowledgements

The authors would like to acknowledge Krisana Kraisintu Foundation for supporting the modified microwave oven in this work. We also acknowledge Prof. Dr. J.E. Moreton, University of Maryland at Baltimore, USA for his assistance in reading and editing English language in the paper.

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