Supplementary Information (SI) for Sustainable Food Technology.

Sustainable Conversion of Waste Robusta Coffee Husk into Tea: Optimization, Properties and Diffusion Kinetics

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Supporting Information

1. Analysis methods

1.1. Moisture Content

Moisture content (w) was determined gravimetrically using an infrared moisture analyzer as the Equation (S1)[1]. Approximately 1 g of homogenized sample was heated at 105 °C until a constant weight was obtained.

$$w = \frac{(m_1 - m_2) \times 100}{m_1} \qquad \text{(S1)}$$

Where m_1 and m_2 are the sample weights before and after drying (g), respectively.

1.2. Antioxidant Activity (DPPH Radical Scavenging Assay)

In the ethanolic extraction, 2 g of RCH sample was soaked in 20 mL of ethanol and shaken at room temperature for 24 h. The mixture was filtered and re-extracted with another 20 mL of ethanol until the filtrate became colorless. Combined ethanolic extracts were collected in Duran glass bottles and stored at 4 °C until analysis[2].

The antioxidant activity (AA) was measured using the DPPH radical method with Trolox as the standard. A 0.002 g DPPH solution was prepared in 100 mL ethanol ($A_{517} \approx 1.10 \pm 0.01$). Trolox stock solution (1000 µM) was serially diluted to 0–750 µM for calibration. Each test tube received 0.1 mL Trolox (or sample) and 4 mL DPPH solution, shaken and incubated for 30 min in the dark at room temperature. Absorbance was measured at 517 nm.

$$\text{RSA (\%)} = \frac{(A_k - A_m) \times 100}{A_k} \qquad \text{(S2)}$$

Where A_k and A_m are the absorbances of the control and sample, respectively. Results were expressed as µmol TE/g dry weight.

1.3. Total Polyphenol Content (TPC)

The ethanolic extract prepared as described in Section 1.2 was used for TPC assay. TPC were quantified using the Folin–Ciocalteu reagent[3]. The reaction mixture contained 0.5 mL diluted sample, 2.5 mL 10% Folin reagent, and after 4 min, 2 mL of 7.5% Na_2CO_3 solution. The mixture was incubated at 40 °C for 30 min and absorbance recorded at 760 nm. A calibration curve was prepared with gallic acid (10–50 mg GAE/L). TPC was expressed as mg GAE/g dry weight.

1.4. Ash Content

Ash content was determined by dry ashing as calculated in Equation (S3)[4]. About 1 g of the dried sample was placed in a pre-weighed crucible and incinerated in a muffle furnace at 525 ± 25 °C until constant weight.

$$Ash\ (\%) = \frac{m_1 \times 100\ \times 100}{m_0 \times S_R} \qquad \text{(S3)}$$

Where m_0 is the initial sample mass, m_1 is the ash mass, and S_R is the sample dry matter (%).

1.5. Reducing Sugars

Reducing sugar content was determined by the 3,5-dinitrosalicylic acid (DNS) colorimetric method[5]. The reaction mixture (1 mL sample + 1 mL DNS) was boiled for 5 min, cooled, diluted with 10 mL distilled water, and the absorbance measured at 540 nm. Glucose (0–1000 µg/mL) served as the standard.

$$G = \frac{X \times V}{m \times N} \qquad \text{(S4)}$$

Where X is glucose concentration (mg/mL) from calibration, V the final volume (mL), N the dilution factor, and m the sample mass (g).

1.6. Total Organic Acids

Total acidity was determined by titration with 0.1 N NaOH using phenolphthalein as the indicator[6].

$$X = \frac{V \times K \times V_2 \times T \times 100}{V_1 \times m} \qquad (S5)$$

Where V is the NaOH volume (mL), K = 0.006 (acetic acid factor), V_1 and V_2 are sample and dilution volumes (mL), m is the sample mass (g), and T = 1.08 (NaOH correction factor).

1.7. Tannin Content

Tannins were determined using the Lowenthal–Neubauer method with $KMnO_4$ as oxidant and indigo carmine as indicator[7]. For titration, 10 mL of the extract, 10 mL of indigo carmine indicator, and 250 mL of distilled water were placed in a 500 mL Erlenmeyer flask and titrated with 0.1 N $KMnO_4$ solution with continuous shaking until a golden-yellow endpoint appeared. A blank titration was performed simultaneously using 10 mL indicator and 250 mL distilled water.

$$X\% = \frac{(a - b) \times V_2 \times 0.00582 \times 100}{V_1 \times m} \qquad (S6)$$

Where a and b are the $KMnO_4$ volumes for sample and blank (mL), V_1 = sample aliquot (10 mL), V_2 = final volume (250 mL), and m = sample mass (g).

1.8. Water-Soluble Extractives

The sample (2 g) were extracted with 200 mL boiling water under gentle reflux for 1 h, filtered, rinsed, and dried at 105 °C for 16 h[8].

$$E(\%) = \frac{((m_0 \times w) - (m_1 \times 100)) \times 100}{m_0 \times w} \qquad (S7)$$

where m_0 is the initial sample mass (g), m_1 is the dry residue (g), and w is the sample dry-matter content (%).

1.9. Caffeine Content

Caffeine was quantified by HPLC method using an RP-18 column with an isocratic mobile phase of 24% methanol in water and UV detection at 272 nm. Extraction was conducted with MgO in hot water (90 °C, 20 min)[9].

$$w_x = \frac{A_x \rho_c V \times 100}{A_c m_s} = \frac{A_x \rho_c \times 25}{A_c m_s} \quad \text{(S8)}$$

where A_x and A_c are the caffeine peak areas of sample and standard, ρ_c is caffeine standard concentration (g/L), V = 0.25 L, and m_s is the sample mass (g).

1.10. Microbial and Ochratoxin A analysis

The quality of the RCH extract was evaluated to determine selected quality parameters in accordance with TCVN 7975:2008 for herbal tea bags and QCVN 8-1:2011/BYT regarding permissible limits of mycotoxin contamination in food products[10]. In brief, microbiological quality was determined using the colony count technique. Samples were serially diluted, and 1 mL aliquots were plated in Petri dishes, overlaid with yeast extract–dextrose–chloramphenicol agar, and incubated aerobically at 25 ± 1 °C for 3–5 days. Plates containing fewer than 150 colonies were counted, and the results were expressed as CFU g^{-1} or CFU mL^{-1} in accordance with the standard.

For detection of Ochratoxin A (OTA), tea was determined using high-performance liquid chromatography (HPLC) with fluorescence detection following immunoaffinity column clean-up. 25 g of RCH tea sample was extracted with methanol–3% sodium bicarbonate solution (1:1, v/v) using high-speed blending. The extract was filtered, diluted with phosphate-buffered saline (PBS), and passed through an OTA-specific immunoaffinity column at a controlled flow rate. After washing with deionized water, OTA was eluted with methanol, evaporated to dryness under a gentle nitrogen stream, and reconstituted with the HPLC mobile phase. Chromatographic separation was achieved on a C18 reversed-phase column using an acetonitrile–methanol–water–

acetic acid mobile phase. Fluorescence detection was performed at excitation and emission wavelengths of 332 and 476 nm, respectively. Quantification was carried out using an external calibration curve with acceptable linearity ($R^2 > 0.99$), and the results were expressed as ng g^{-1} of sample.

2. Optimization of Robusta coffee husk Roasting

Before roasting, the raw RCH was analyzed for its proximate composition, including moisture, carbohydrates, proteins, lipids, ash, antioxidant activity, TPC and organic acids. As shown in Table S5, carbohydrates represented the dominant fraction of the dry matter, while proteins and reducing sugars served as key nitrogen- and carbon-based precursors for Maillard and caramelization reactions during subsequent thermal processing. The dried husk contained protein (10.38 g/100 g dw), reducing sugars (13.48 ± 0.23 g/100 g dw), lipid (1.18 g/100 g dw), ash (5.43 ± 0.12 g/100 g dw), and total organic acids (1.08 ± 0.04 g/100 g dw), with a residual moisture content of 13.57 ± 0.18 g/100 g dw. The high levels of simple sugars, organic acids, and minor lipid fractions provided essential substrates for browning and flavor formation, whereas the relatively elevated ash and protein contents indicated substantial mineral and nitrogen reserves within the outer husk layers.

The 90% ethanolic extract of the dried RCH exhibited TPC of 289.24 ± 3.73 mg GAE/g dry weight and AA of 1348.61 ± 17.23 µmol TE/g dry weight. In comparison, the aqueous extract accounted for only 16.71% of the TPC and 39.85% of the AA relative to the exhaustive ethanolic extract, confirming that phenolic constituents were primarily responsible for DPPH radical scavenging activity. These compositional characteristics indicate that raw coffee husks possess abundant biochemical precursors and antioxidant compounds, making them promising raw materials for roasting optimization and instant-tea development.

The roasting process of coffee husk was optimized using a central composite design (CCD) within the framework of response surface methodology, considering roasting temperature (X_1, °C) and roasting time (X_2, s) as independent variables and five responses as targets: total polyphenols (Y_1), antioxidant activity (Y_2), reducing sugars (Y_3), total organic acids (Y_4), and tannins (Y_5). The experimental and model-predicted values are summarized in Tables S6 and S7. The close agreement between observed and predicted responses across all design points indicates good predictive ability of the fitted models.

All responses were adequately described by second-order polynomial equations including linear, quadratic, and interaction terms of X_1 and X_2 (Table S8). The RSM models showed non-significant lack-of-fit, as evidenced by F values lower than the corresponding critical F values at the 95% confidence level ($F < F_{0.05}$), confirming the adequacy of the proposed models. In addition, the high R^2 values (0.9944–0.9999) with the close agreement between adjusted R^2 and predicted R^2 indicated that the models are not overfitted and exhibit excellent predictive capability. These results demonstrated that the developed regression models were statistically reliable and suitable for optimization and prediction within the roasting conditions.

The response surface analysis (Figure S1) demonstrated that both roasting temperature and time exerted significant and interactive significantly effects on all quality parameters of the coffee husk. The trends of these responses were characterized by initial increases followed by subsequent declines under severe roasting conditions. At moderate roasting conditions (around 115–120 °C and 220–230 s), both TPC and AA reached their maximum levels. This indicated the release and transformation of phenolic and non-phenolic compounds into lower-molecular-weight forms with higher antioxidant potential. This enhancement likely arises from the thermal degradation of chlorogenic acid and the formation of simpler phenolic acids such as caffeic, ferulic, and *p*-coumaric acids (Table S9).

Temperature and prolonged exposure induced decomposition and polymerization of phenolic compounds into high-molecular weight melanoidin structures that can lead to a gradual decrease in both TPC and DPPH radical scavenging capacity [11]. Moreover, transformation of non-phenolic compounds such as aminoacid molecules could also create the Maillard reaction products with high AA [12, 13]. At the optimal roasting point (117 °C, 225 s), total polyphenols and AA reached 39.22 ± 0.14 mg GAE/g and 499.93 ± 4.25 µmol TE/g dw, respectively.

Reducing sugar content exhibited a continuous decline with increasing roasting intensity. This decrease is attributed to the participation of reducing sugars in Maillard and caramelization reactions, in which amino acids react with carbonyl compounds to form nitrogenous polymers (melanoidins) responsible for the brown color and characteristic aroma of roasted husk [14]. The moderate loss of sugars at 117 °C for 225 s (7.27 ± 0.04 g/100 g dw) indicates that these conditions favor the generation of flavor-active intermediates while avoiding excessive thermal degradation. Moreover, the maximum total organic acid level (0.82 ± 0.01 g/100g dw) was observed near the same optimal temperature – time range as for polyphenols, suggesting coupled pathways between phenolic degradation and organic acid formation.

Similarly, tannin content increased progressively with temperature and time until reaching a plateau near the optimum. The maximum tannin content was recorded at 4.48 ± 0.14 g/100g dw %, corresponding to 117 °C and 225 s. Therefore, both roasting temperature and time exhibited strong synergistic effects on the compositional and functional characteristics of coffee husk. The identified optimum (117 °C for 225 s) thus represents a balanced condition, maximizing phenolic and antioxidant attributes while maintaining desirable chemical and sensory qualities.

3. Optimization of aqueous extraction of roasted RCH

The aqueous extraction process of roasted RCH was optimized using a Box–Behnken design (BBD) within the framework of response surface methodology (RSM). Three independent variables were selected: solvent-to-husk ratio (X_1, v/w), extraction temperature (X_2, °C), and extraction time (X_3, min). The optimization targeted five responses representing the main quality indicators of the extract: total polyphenols (TPC, Y_1, mg GAE g^{-1} dw), antioxidant activity (AA, Y_2, μmol TE g^{-1} dw), reducing sugars (Y_3, g 100 g^{-1} dw), total organic acids (Y_4, g/100g dw), and tannins (Y_5, g/100g dw). The experimental and model-predicted results are summarized in Tables S10, S11, and S12. The close agreement between observed and predicted values across the design space indicates that the regression models adequately describe the experimental data and possess strong predictive ability.

Response surface analysis (Figures S2–S6) demonstrated that the solvent-to-husk ratio, extraction temperature, and extraction time exhibited significant linear, quadratic, and interactive effects on all five response variables. The responses followed non-linear behaviors characterized by an initial increase followed by a subsequent decline under excessive extraction conditions. Moderate extraction parameters, specifically a solvent-to-husk ratio of approximately 35:1, temperature near 70 °C, and extraction time of about 45 min, produced the highest TPC and AA values, suggesting that controlled thermal energy and adequate solvent diffusion facilitated the liberation. These observations highlighted the need to balance solvent volume, temperature, and duration to maximize extraction yield while preventing degradation of thermolabile compounds.

Among the studied parameters, extraction temperature exerted the most pronounced effect on solute mobility and matrix permeability. Increasing temperature from 50 °C to 70 °C significantly enhanced both TPC and AA due to improved solvent penetration and partial disruption of the lignocellulosic structure. However, at temperatures exceeding 75 °C, thermal degradation and oxidation of phenolic components occurred, resulting in a decrease in both responses. The maximum TPC (42.11 mg GAE g^{-1} dw) and AA (525.55 µmol TE g^{-1} dw) were obtained at 70 °C. Similarly, total organic acids and tannins exhibited parabolic trends, reaching respective maxima of 0.91 g/100g dw and 4.58 g/100g dw. The tannin content of instant tea products is a critical factor influencing their sensory properties. Although tannins contribute to antioxidant activity, excessive levels are known to impart bitterness and astringency. In the present study, the optimized tannin content was maintained at a moderate level, which did not adversely affect the flavor profile of the instant tea while allowing for the maximization of total polyphenol content and antioxidant capacity. This balance between total polyphenols and tannins is essential for instant tea applications, where both biofunctional value and palatability must be carefully considered. Accordingly, the selected extraction conditions are expected to provide an appropriate compromise between antioxidant enrichment and acceptable sensory quality.

Extraction time showed a synergistic interaction with temperature and solvent ratio. Prolonging extraction improved mass transfer and promoted swelling of cell walls, enhancing solute diffusion and polyphenol release. However, excessive extraction time (> 50 min) led to oxidative degradation of heat-sensitive compounds. The optimal duration of 45 min yielded maximum polyphenol and antioxidant responses, confirming its suitability for maintaining equilibrium between diffusion efficiency and structural stability.

Comprehensive multi-response optimization using the desirability function identified an optimal region corresponding to a solvent-to-husk ratio of 35.807, extraction temperature of 69.7 °C, and extraction time of 44.5 min. Under these conditions, the model predicted TPC = 42.107 mg GAE g^{-1}, AA = 525.551 µmol TE g^{-1}, reducing sugars = 7.429 g/100g dw, total organic acids

= 0.906 g/100g dw, and tannins = 4.584 g/100g dw. Validation experiments performed under practical conditions (solvent-to-husk ratio of 36, temperature of 70 ± 2 °C, and extraction time of 45 min) yielded experimental values of 40.31 ± 0.66 mg GAE g^{-1}, 518.24 ± 4.83 µmol TE g^{-1}, 7.13 ± 0.19 g/100g dw, 0.85 ± 0.02 g/100g dw, and 4.47 ± 0.11 g/100g dw for Y_1–Y_5, respectively. The minimal deviations between predicted and experimental results confirmed the robustness and reliability of the BBD-derived models, validating the optimized conditions as highly effective for maximizing the recovery of phenolic compounds and antioxidant capacity from RCH using water as a sustainable solvent.

4. Tables

Table S1. Experimental factors and coded levels used in the RSM – CCD design for Robusta coffee husk roasting.

Factor	**Unit**	**Code**	**Range**	**− α**	**−1**	**0**	**+1**	**+α**
Roasting temperature	°C	X_1	100–180	83	100	140	180	197
Roasting time	s	X_2	150–270	125	150	210	270	295

Table S2. Experimental matrix of the central composite design for optimizing Robusta coffee husk roasting conditions.

Run	**X_1 (coded)**	**X_2 (coded)**
1	0	−α
2	−1	1
3	0	0
4	0	0
5	−1	−1
6	0	+α
7	1	−1
8	0	0
9	0	0
10	+α	0
11	−α	0

Run	**X_1 (coded)**	**X_2 (coded)**
12	1	1
13	0	0

Table S3. Experimental factors and coded levels used in the RSM–Box–Behnken design for aqueous extraction of Robusta coffee husk.

Factor	**Unit**	**Code**	**Range**	**−1**	**0**	**+1**
Solvent-to-husk ratio	mL/g	X_1	10–50	10	30	50
Extraction temperature	°C	X_2	50–90	50	70	90
Extraction time	min	X_3	10–70	10	40	70

Table S4. Experimental matrix of the RSM–Box–Behnken design for aqueous extraction of coffee husk.

Run	**X_1 (coded)**	**X_2 (coded)**	**X_3 (coded)**
1	0	−1	1
2	0	0	0
3	0	1	−1
4	0	−1	−1
5	0	0	0
6	0	0	0
7	−1	1	0
8	0	1	1
9	0	0	0
10	1	1	0
11	1	−1	0
12	−1	0	1
13	−1	−1	0
14	0	0	0
15	1	0	1
16	−1	0	−1
17	1	0	−1

Table S5. Properties of dried raw coffee husk

Component	**Content**
Moisture (g/100g dw)	13.57 ± 0.18
Protein (g/100g dw)	10.38 ± 0.38
Lipid (g/100g dw)	1.18 ± 0.15
Ash (g/100g dw)	5.43 ± 0.12
Reducing sugars (g/100g dw)	13.48 ± 0.23

Component	**Content**
Total organic acids (g/100g dw)	1.08 ± 0.04
TPC (mg GAE/g dry weight)	
Ethanolic extract	289.24 ± 3.73
Water extract	48.32 ± 0.63
DPPH (µmol TE/g dry weight)	
Ethanolic extract	1348.61 ± 17.23
Water extract	537.45 ± 8.57

Table S6. Predicted results of the Central Composite Design (CCD) model

No.	X_1	X_2	Y_1	Y_2	Y_3	Y_4	Y_5
1	0	$-\alpha$	32.34	475.98	7.26	0.73	3.836
2	-1	1	37.86	476.55	6.64	0.93	4.490
3	0	0	39.04	498.65	6.53	0.84	4.480
4	0	0	39.04	498.65	6.53	0.84	4.480
5	-1	-1	28.58	377.79	8.29	0.67	3.255
6	0	$+\alpha$	32.34	384.84	3.30	0.73	3.836
7	1	-1	33.74	466.57	5.30	0.65	4.011
8	0	0	39.04	498.65	6.53	0.84	4.480
9	0	0	39.04	498.65	6.53	0.84	4.480
10	$+\alpha$	0	27.07	296.89	2.58	0.39	3.092
11	$-\alpha$	0	32.89	402.13	8.43	0.79	3.770
12	1	1	24.46	238.93	1.35	0.39	2.776
13	0	0	39.04	498.65	6.53	0.84	4.480

Table S7. Experimental results of the Central Composite Design (CCD) model

No.	X_1	X_2	Y_1	Y_2	Y_3	Y_4	Y_5
1	0	$-\alpha$	32.51	476.03	7.28	0.75	3.825
2	-1	1	38.12	478.44	6.65	0.93	4.525
3	0	0	39.26	498.54	6.54	0.85	4.489
4	0	0	38.85	497.08	6.55	0.83	4.491
5	-1	-1	28.82	378.94	8.28	0.67	3.267
6	0	$+\alpha$	31.93	383.72	3.32	0.72	3.828
7	1	-1	33.67	465.28	5.27	0.63	3.998
8	0	0	39.26	502.45	6.51	0.84	4.488
9	0	0	38.98	498.27	6.54	0.83	4.490
10	$+\alpha$	0	27.03	295.63	2.58	0.41	3.084
11	$-\alpha$	0	32.47	398.23	8.44	0.79	3.732
12	1	1	24.42	238.36	1.33	0.37	2.787
13	0	0	38.87	496.89	6.53	0.85	4.452

Table S8. Regression equations for the roasting optimization model

Response (Y)	**Regression Equation**	**Model Fit (R^2, F)**
Y_1 – Total polyphenols (mg GAE/g dw)	$Y_1 = 39.04 - 2.06X_1 - 4.64X_1X_2 - 4.53X_1^2 - 3.35X_2^2$	$R^2 = 0.9984$; Adj. $R^2 = 0.9972$; Pre. $R^2 = 0.9913$ $F = 4.11 < F_{0.05;3,4} = 6.59$
Y_2 – Antioxidant activity (µmol TE/g dw)	$Y_2 = 498.65 - 37.21X_1 - 32.22X_2 - 81.60X_1X_2 - 74.57X_1^2 - 34.12X_2^2$	$R^2 = 0.9996$; Adj. $R^2 = 0.9994$; Pre. $R^2 = 0.9986$

		$F = 2.47 < F_{0.05;2,4} = 6.94$
Y_3 – Reducing sugars (g/100 g dw)	$Y_3 = 6.53 - 2.07X_1 - 1.40X_2 - 0.58X_1X_2 - 0.51X_1^2 - 0.62X_2^2$	$R^2 = 0.9999$; Adj. $R^2 = 0.9999$; Pre. $R^2 = 0.9995$ $F = 4.05 < F_{0.05;2,4} = 6.94$
Y_4 – Total organic acids (g/100 g dw)	$Y_4 = 0.84 - 0.14X_1 - 0.13X_1X_2 - 0.12X_1^2 - 0.06X_2^2$	$R^2 = 0.9944$; Adj. $R^2 = 0.9905$; Pre. $R^2 = 0.9667$ $F = 5.26 < F_{0.05;3,4} = 6.59$
Y_5 – Tannins (g/100 g dw)	$Y_5 = 4.48 - 0.24X_1 - 0.62X_1X_2 - 0.52X_1^2 - 0.32X_2^2$	$R^2 = 0.9993$; Adj. $R^2 = 0.9988$; Pre. $R^2 = 0.9962$ $F = 3.87 < F_{0.05;3,4} = 6.59$

Table S9. Chemical compositions of roasted coffee husk

Component	**Content**
Carbohydrates (g/100g dw)	57.8 ± 1.64
Moisture (g/100g dw)	12.0 ± 3.2
Lipids (g/100g dw)	1.7 ± 0.34
Total fiber (g/100g dw)	31.9 ± 0.42
Ash (g/100g dw)	6.0 ± 0.08
Protein (g/100g dw)	9.2 ± 0.73
Caffeine (g/100g dw)	1.2 ± 0.21
Reducing sugar (g/100g dw)	7.27 ± 0.04
Tannins (g/100g dw)	4.48 ± 0.13
Total organic acid (g/100g dw)	0.82 ± 0.01
Chlorogenic acid (g/100g dw)	12.6 ± 0.07
TPC (water extract, mgGAE/g dried weight)	39.22 ± 0.14
AA (water extract, mgGAE/g dried weight)	499.93 ± 4.25

Table S10. Predicted extraction results of the Box–Behnken Design (BBD) model

No.	X_1	X_2	X_3	Y_1	Y_2	Y_3	Y_4	Y_5
1	0	-1	1	39.11	505.22	7.44	0.81	4.345
2	0	0	0	41.99	524.45	7.42	0.91	4.580
3	0	1	-1	38.55	495.76	6.96	0.85	4.257
4	0	-1	-1	38.11	495.76	6.96	0.85	4.257
5	0	0	0	41.99	524.45	7.42	0.91	4.580
6	0	0	0	41.99	524.45	7.42	0.91	4.580
7	-1	1	0	38.93	501.46	6.96	0.83	4.358
8	0	1	1	39.55	505.22	6.72	0.81	4.345
9	0	0	0	41.99	524.45	7.42	0.91	4.580
10	1	1	0	40.27	508.12	6.96	0.83	4.358

11	1	-1	0	39.38	508.12	7.32	0.83	4.358
12	-1	0	1	40.19	512.69	7.36	0.89	4.443
13	-1	-1	0	38.96	501.46	7.32	0.83	4.358
14	0	0	0	41.99	524.45	7.42	0.91	4.580
15	1	0	1	41.07	519.35	7.36	0.89	4.443
16	-1	0	-1	39.19	503.23	7.24	0.93	4.355
17	1	0	-1	40.06	509.89	7.24	0.93	4.355

Table S11. Experimental extraction results of the Box–Behnken Design (BBD) model

No.	**X_1**	**X_2**	**X_3**	**Y_1**	**Y_2**	**Y_3**	**Y_4**	**Y_5**
1	0	-1	1	39.12	503.05	7.42	0.82	4.347
2	0	0	0	42.05	525.04	7.43	0.92	4.643
3	0	1	-1	38.64	497.05	6.97	0.84	4.289
4	0	-1	-1	38.12	497.24	6.96	0.84	4.246
5	0	0	0	41.88	522.97	7.41	0.89	4.563
6	0	0	0	41.96	524.82	7.43	0.90	4.521
7	-1	1	0	38.98	501.45	6.96	0.81	4.296
8	0	1	1	39.42	504.61	6.72	0.82	4.328
9	0	0	0	42.13	525.65	7.41	0.92	4.587
10	1	1	0	40.24	511.09	6.94	0.84	4.483
11	1	-1	0	39.32	506.44	7.33	0.83	4.335
12	-1	0	1	40.08	514.22	7.36	0.87	4.483
13	-1	-1	0	38.98	500.18	7.32	0.82	4.322
14	0	0	0	41.91	523.76	7.42	0.91	4.592
15	1	0	1	41.28	520.56	7.36	0.88	4.423
16	-1	0	-1	39.21	502.98	7.22	0.93	4.345
17	1	0	-1	39.92	507.37	7.23	0.92	4.349

Table S12. Regression equations for the aqueous extraction optimization model

Response (Y)	**Regression equation**	**Model fit (R², F)**
Y_1 – Total polyphenols (mg GAE/g dw)	$Y_1 = 41.99 + 0.4388X_1 + 0.2175X_2 + 0.5012X_3 + 0.23X_1X_2 - 0.6542X_1^2 - 1.95X_2^2 - 1.21X_3^2$	$R^2 = 0.9973$; Adj. $R^2 = 0.9939$; Pre. $R^2 = 0.9776$ $F = 2.61 < F_{0.05;4,4} = 6.39$
Y_2 – Antioxidant activity (µmol TE/g dw)	$Y_2 = 524.45 + 3.33X_1 + 4.73X_3 - 4.43X_1$^2 $- 15.23X_2^2 - 8.73X_3^2$	$R^2 = 0.9857$; Adj. $R^2 = 0.9673$; Pre. $R^2 = 0.8079$ $F = 4.71 < F_{0.05;6,4} = 6.16$
Y_3 – Reducing sugars	$Y_3 = 7.42 - 0.18X_2 + 0.06X_3 -$	$R^2 = 0.9984$; Adj. $R^2 =$

(g/100 g dw)	$0.1775X_2X_3 - 0.2787X_2^2 - 0.1238X_3^2$	0.9963; Pre. $R^2 = 0.9809$ $F = 2.19 < F_{0.05;6,4} = 6.16$
Y_4 – Total organic acids (g/100 g dw)	$Y_4 = 0.908 - 0.0175X_3 - 0.0765X_2^2$	$R^2 = 0.9347$; Adj. $R^2 = 0.9254$; Pre. $R^2 = 0.9062$ $F = 0.97 < F_{0.05;\,9,4} = 5.91$
Y_5 – Tannins (g/100 g dw)	$Y_5 = 4.58 + 0.044X_3 - 0.0624X_1^2 - 0.1598X_2^2 - 0.1189X_3^2$	$R^2 = 0.8688$; Adj. $R^2 = 0.8251$; Pre. $R^2 = 0.7625$ $F = 1.79 < F_{0.05;7,4} = 6.09$

Table S13. Physicochemical and microbiological properties of the Roasted RCH Extract under optimal extraction conditions

Parameters	**Content**
Reducing sugars (g/100g dw)	71.3 ± 1.9
Total organic acids (g/100g dw)	8.5 ± 0.2
Soluble solids (g/100g dw)	365.8 ± 26.0
Caffeine (g/100g dw)	9.2 ± 0.6
Total polyphenols (mg GAE/g dw)	40.31 ± 0.66
Antioxidant activity (µmol TE/g dw)	518.24 ± 4.83
Tannins (g/100g dw)	44.7 ± 1.1
Total yeast and mold count (CFU/g)	Not detected (LOD = 10)
Total aerobic microorganisms (CFU/g)	1.0×10^5
Ochratoxin A (µg/kg)	Not detected (LOD = 0.30)

Note: Values are expressed as mean ± standard deviation (n = 3). LOD: limit of detection

Table S14. Scoring Criteria according to *TCVN 3215-79* for instant RCH tea

Attribute	**Score**	**Description**
Color	5	Very natural color, highly characteristic of the product
	4	Natural color, characteristic of the product
	3	Relatively natural color, moderately characteristic
	2	Slightly discolored, not characteristic
Aroma–Taste	5	Very characteristic coffee husk tea aroma and taste
	4	Characteristic coffee husk tea aroma and taste
	3	Moderately characteristic coffee husk tea aroma and taste
	2	Non-characteristic coffee husk tea aroma and taste

Appearance	5	Very clear solution, completely dissolved, homogeneous
	4	Clear solution, well dissolved, homogeneous
	3	Relatively clear, dissolved but slightly non-homogeneous
	2	Turbid solution, uneven dissolution, non-homogeneous

Table S15. Sensory evaluation results of instant coffee husk tea

Panelist	**Color**	**Aroma–Taste**	**Appearance**
1	4	4	5
2	4	4	5
3	5	4	5
4	5	4	5
5	5	5	5
6	5	5	5
7	4	5	5
8	4	4	5
9	5	5	5
10	4	4	4
11	5	5	5
12	4	5	4
13	5	5	5
14	4	5	5
15	5	5	5
16	4	5	4
17	5	5	5
18	4	5	5
19	5	5	5
20	5	5	5
Total scores	91	94	97
Average score (unweighted)*	4.55	4.70	4.85
Weighting factors	1.4	1.4	1.2
Weighted average scores	6.37	6.58	5.82
Overall quality score	18.77		

**According to TCVN 3215-79: a product is classified as "Good" if the unweighted average score for each attribute is ≥ 4.8; a product is classified as "Fair" if the unweighted average* *score for each attribute is ≥ 3.8.*

5. Figures

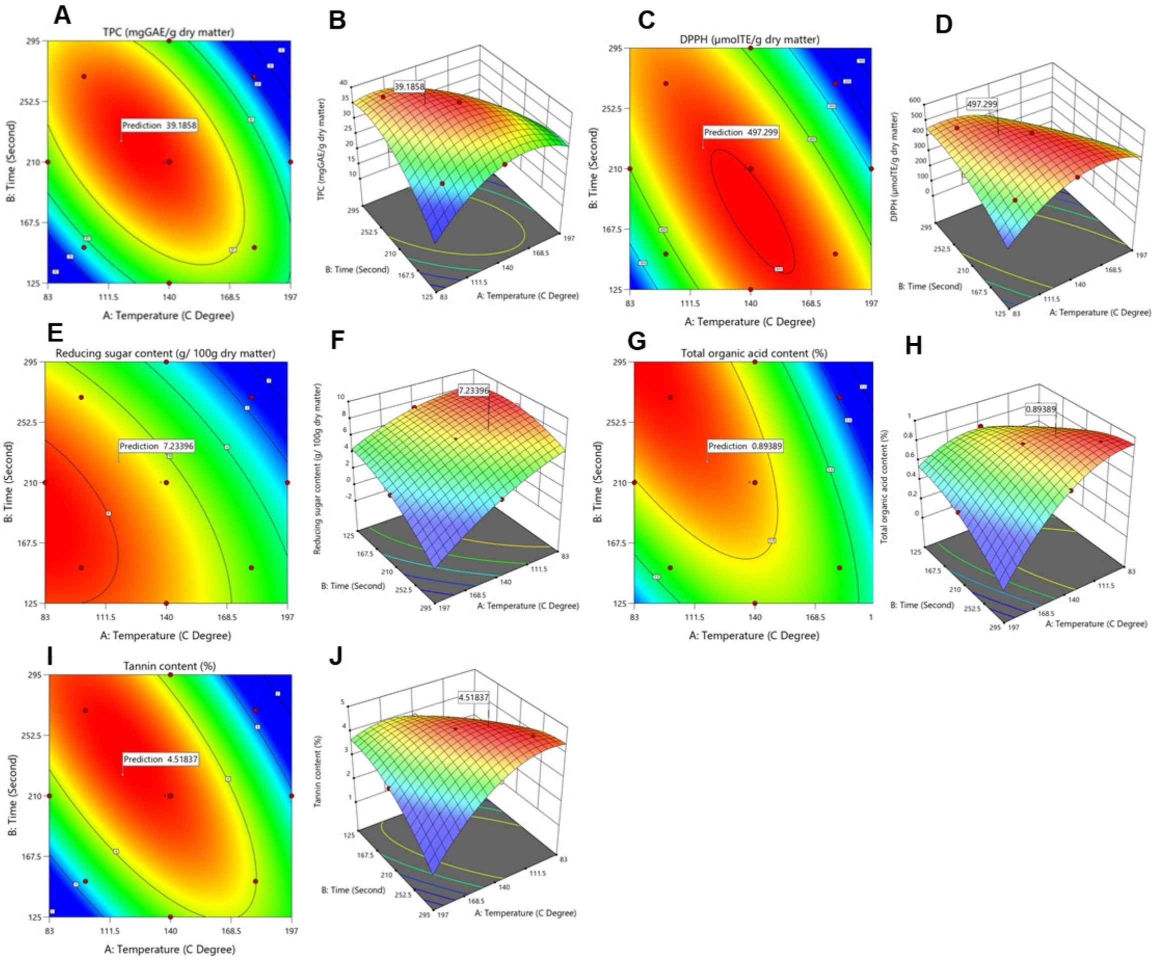

Figure S1. Two-dimensional (2D) and three-dimensional (3D) response surface plots illustrating the effects of roasting temperature (°C) and roasting time (s) on the functional properties of Robusta coffee husk: (A, B) total polyphenol content (TPC), (C, D) antioxidant activity, (E, F) reducing sugar content, (G, H) total organic acids, and (I, J) tannin content. Most responses exhibited an initial increase followed by a decline under severe roasting, with optimal phenolic and antioxidant characteristics observed at moderate conditions (around 117 °C and 225 s).

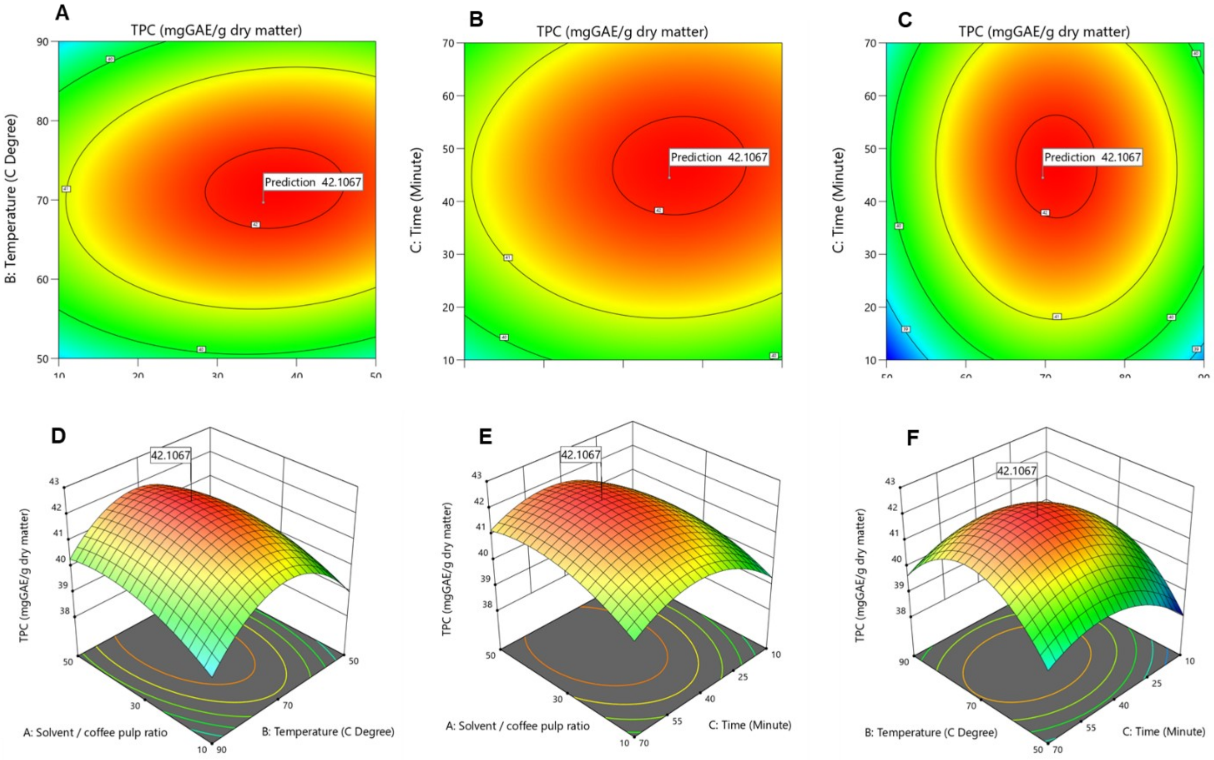

Figure S2. Two-dimensional (2D) and three-dimensional (3D) response surface plots showing the effects of solvent-to-husk ratio (A, B), extraction temperature (C, D), and extraction time (E, F) on the total polyphenol content.

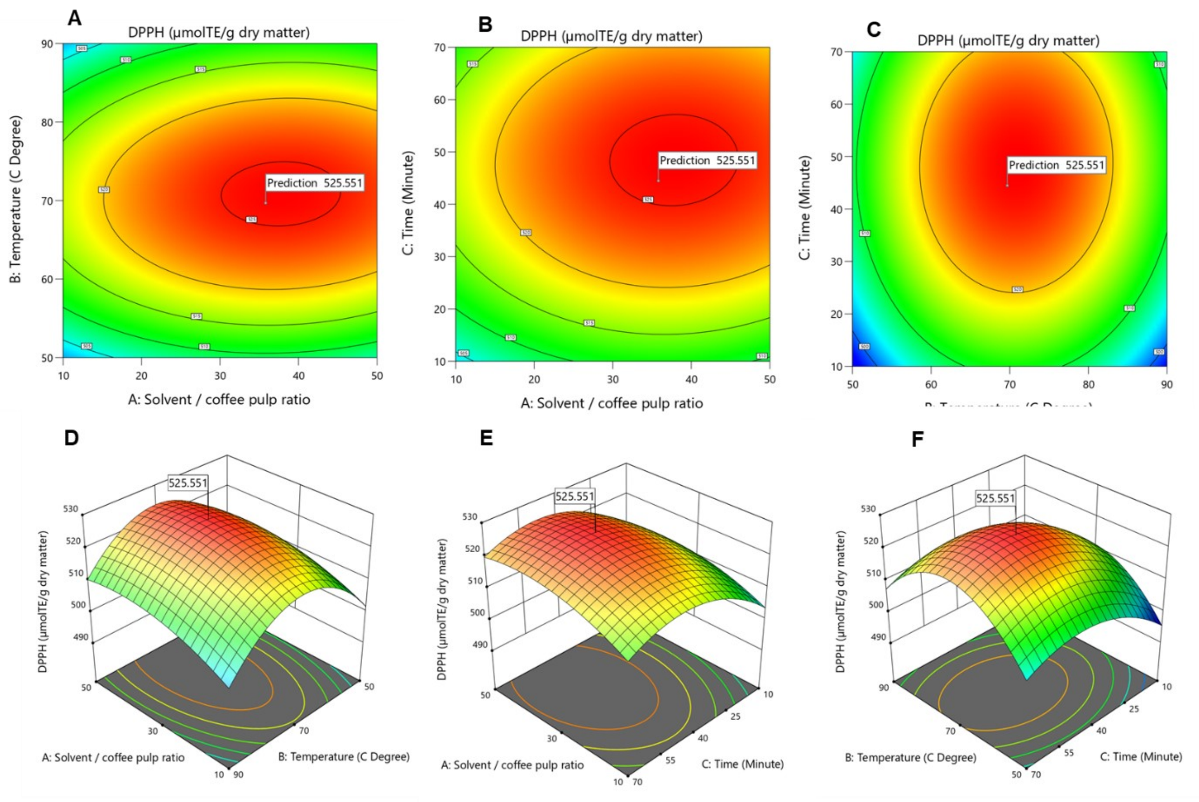

Figure S3. Two-dimensional (2D) and three-dimensional (3D) response surface plots showing the effects of solvent-to-husk ratio (A, B), extraction temperature (C, D), and extraction time (E, F) on the antioxidant activity.

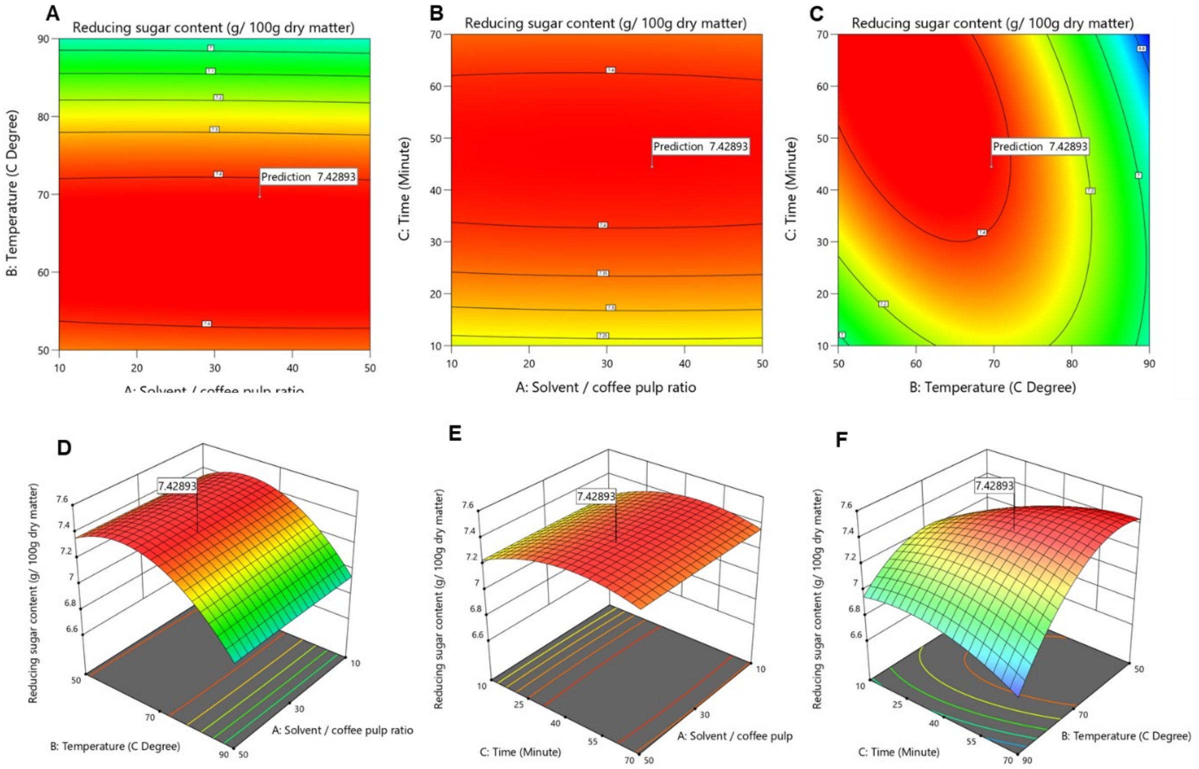

Figure S4. Two-dimensional (2D) and three-dimensional (3D) response surface plots showing the effects of solvent-to-husk ratio (A, B), extraction temperature (C, D), and extraction time (E, F) on the reducing sugar.

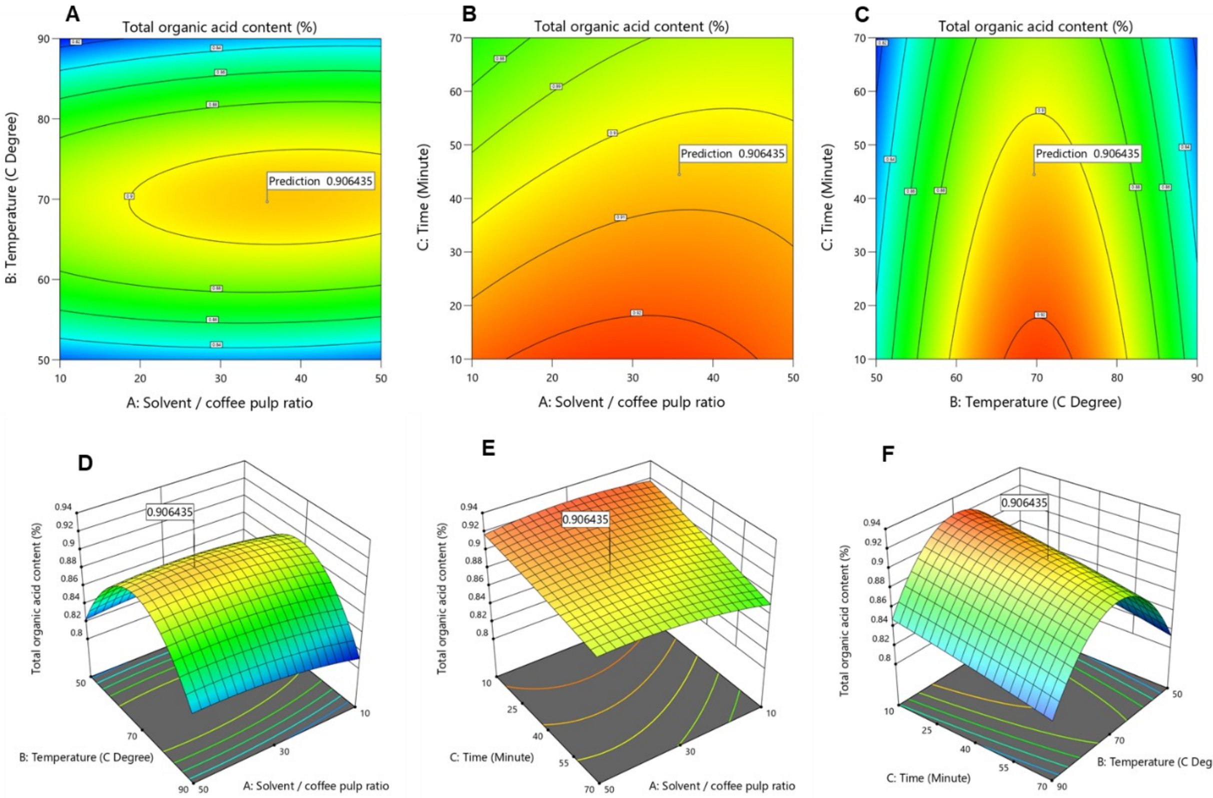

Figure S5. Two-dimensional (2D) and three-dimensional (3D) response surface plots showing the effects of solvent-to-husk ratio (A, B), extraction temperature (C, D), and extraction time (E, F) on the total organic acids.

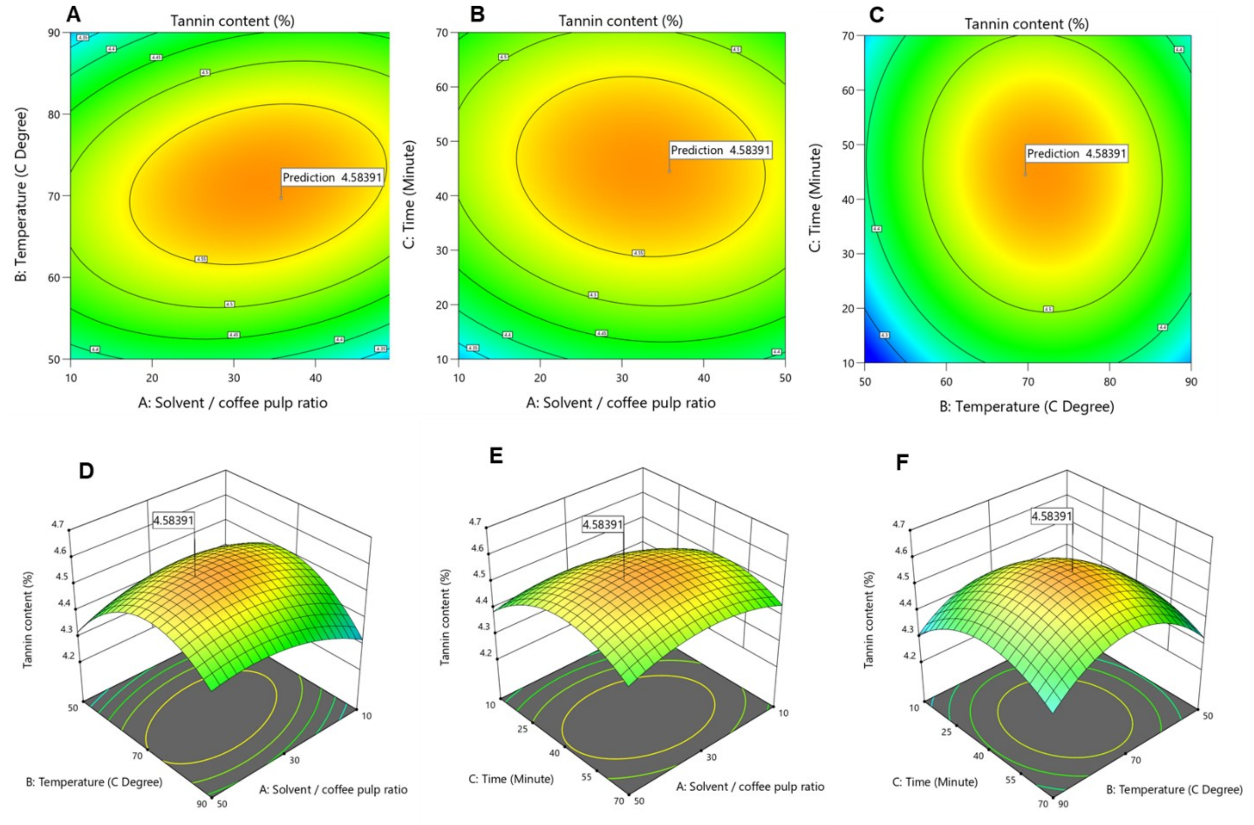

Figure S6. Two-dimensional (2D) and three-dimensional (3D) response surface plots showing the effects of solvent-to-husk ratio (A1, A2), extraction temperature (B1, B2), and extraction time (C1, C2) on the tannin content.

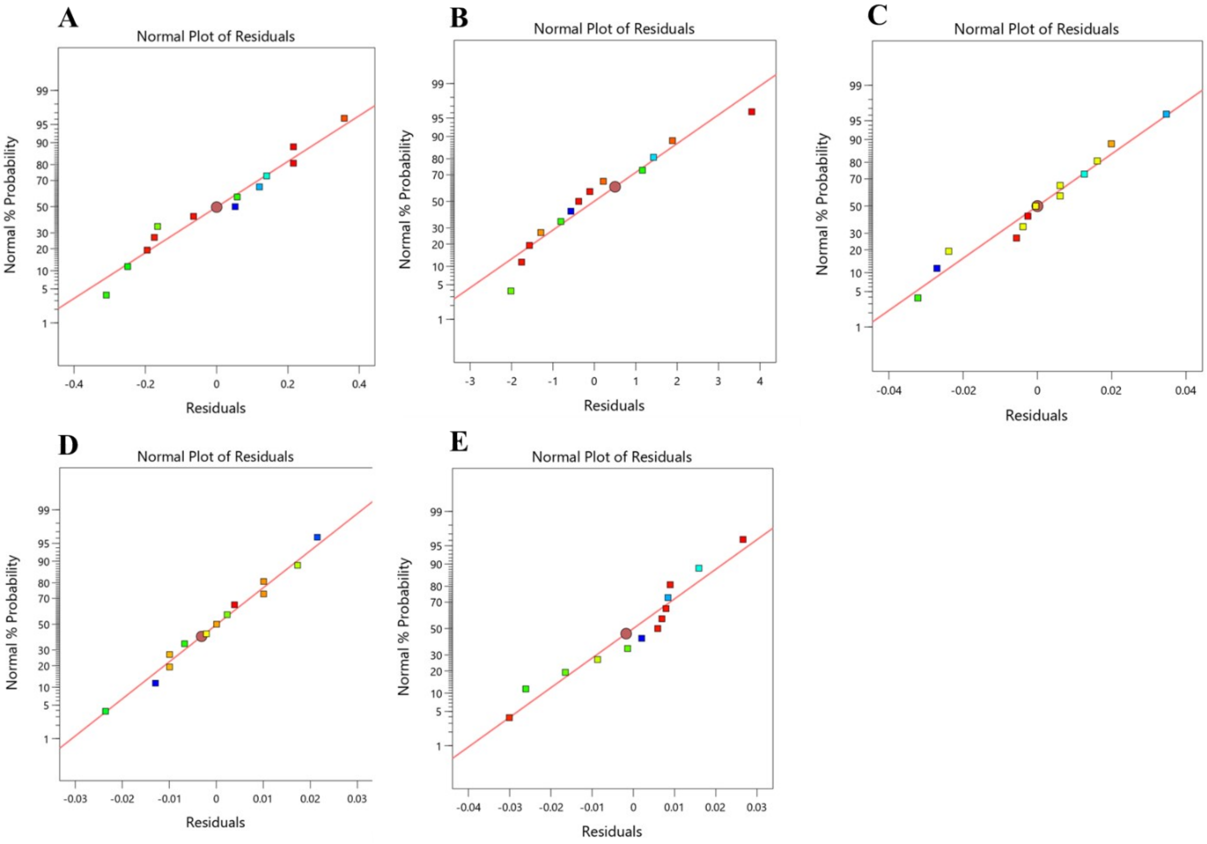

Figure S7. Normal probability plot of residuals for the prediction model of RCH roasting process: (A) TPC; (B) DPPH; (C) Reducing sugar: (D) Total organic acid; and (E) Tannin content.

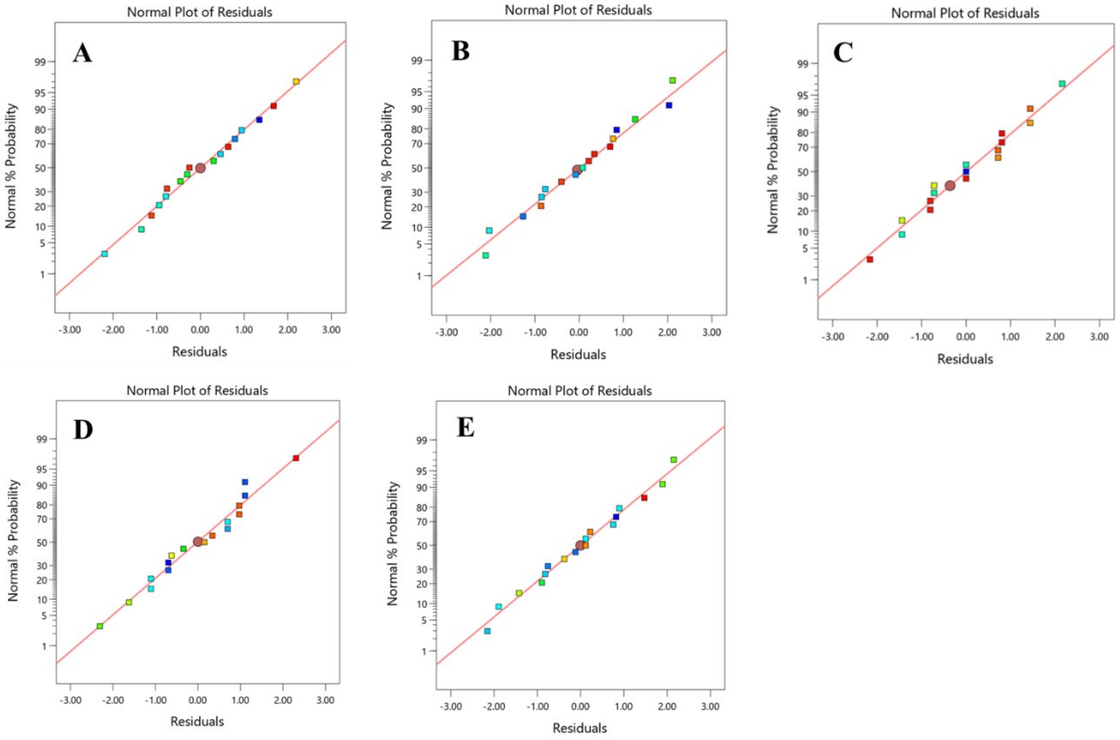

Figure S8. Normal probability plot of residuals for the prediction model of extraction process of roasted RCH: (A) TPC; (B) DPPH; (C) Reducing sugar: (D) Total organic acid; and (E) Tannin content.

1. D. Camuffo, *Journal of Cultural Heritage*, 2018, **31**, S10-S14.
2. İ. Gulcin and S. H. Alwasel, *Processes*, 2023, **11**, 2248.
3. N. Turkmen, Y. S. Velioglu, F. Sari and G. Polat, *Molecules*, 2007, **12**, 484-496.
4. L. Gruber, L. Seidl, M. Zanetti and T. Schnabel, *Forests*, 2021, **12**, 1480.
5. N. N. Deshavath, G. Mukherjee, V. V. Goud, V. D. Veeranki and C. V. Sastri, *International journal of biological macromolecules*, 2020, **156**, 180-185.
6. V. Paul, A. Singh and R. Pandey, *Post-harvest physiology of fruits and flowers*, 2010, **44**.
7. D. Brand, A. Pandey, S. Roussos and C. R. Soccol, *Enzyme and microbial technology*, 2000, **27**, 127-133.
8. W. Sui, Y. Xiao, R. Liu, T. Wu and M. Zhang, *Journal of Food Engineering*, 2019, **261**, 51-59.
9. E. Nishitani and Y. M. Sagesaka, *Journal of Food Composition and Analysis*, 2004, **17**, 675-685.

10. L. Shang, X. Bai, C. Chen, L. Liu, M. Li, X. Xia and Y. Wang, *Food Control*, 2019, **106**, 106743.
11. D. Perrone, A. Farah and C. M. Donangelo, *Journal of agricultural and food chemistry*, 2012, **60**, 4265-4275.
12. L. N. Vhangani and J. Van Wyk, *Food Chemistry*, 2016, **208**, 301-308.
13. F. J. Morales and S. Jiménez-Pérez, *Food chemistry*, 2001, **72**, 119-125.
14. K. Brudzynski and D. Miotto, *Food Chemistry*, 2011, **127**, 1023-1030.