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Anthocyanin and Ascorbic Acid Degradation in Sonicated Strawberry Juice

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Strawberry juice samples were sonicated at amplitude levels ranging from 40 to 100% at a constant frequency of 20 kHz for treatment times (2−10 min) and pulse durations of 5 s on and 5 s off. Sonication was found to reduce anthocyanin and ascorbic acid contents by 3.2 and 11%, respectively, at the maximum treatment conditions. Response surface methodology (RSM) based on a two-factor, five-level central composite design was employed to determine the effect of amplitude level and treatment time on anthocyanins (P3G), ascorbic acid (AA) content, and color values (L^*, a^*, and b^*). The model predictions for the selected nutritional and quality parameters were closely correlated to the experimental results. RSM was demonstrated to be an effective technique to model the effect of sonication on strawberry juice quality while minimizing the number of experiments required.

KEYWORDS: Sonication; color degradation; ascorbic acid; anthocyanin; strawberry juice

INTRODUCTION

Strawberries (*Fragaria* × *ananassa* Duch.) have high levels of micronutrients and phytochemical compounds (*1*), particularly anthocyanins, vitamin C, and phenolic compounds (*2*). The levels of these phytochemicals strongly influence the sensorial-organoleptic attributes and nutritional value. Pelargonidin-3-glucoside (P3G) is the major anthocyanin found in strawberries (**Figure 1**) and is responsible for the attractive, bright red color of fresh strawberries (*3*). Anthocyanins provide an array of health-promoting benefits (*4*), are known for their pharmacological properties, and are employed for therapeutic purposes (*5*). Factors influencing anthocyanins stability include pH, light, oxygen, enzymes such as polyphenols oxidases, ascorbic acid, and high temperature during processing and storage (*6, 7*). The stability of anthocyanins is also influenced by other fruit components, particularly the interaction of ascorbic acid with anthocyanin, which has been reported in various fruit juice model systems including cranberry juice (*8*) and strawberry and black currant products (*9*). The interaction of ascorbic acid with anthocyanin pigments results in the mutual degradation and a decrease in color (*10*). This is also reported for strawberry juice (*11*).

Power ultrasound is a novel and emerging process technology that has been investigated for a range of processing operations (*12, 13*). Power ultrasound has been reported to be effective against foodborne pathogens in orange juice (*14*), apple cider and milk (*15*), guava juice in combination with carbonation (*16*), and water cress in combination with heat (*17*). Sonication is a promising alternative to conventional thermal processing and has been identified as a potential technology to meet the FDA requirement of a 5 log reduction in pertinent microorganisms found in fruit juices (*18*). Previous work has demonstrated that sonication has a minimal effect on ascorbic acid (*19*) and enhances orange juice cloud value (*20*).

A number of studies have investigated the thermal degradation of anthocyanins for blackberry (*21*), sour cherry (*22*), raspberry (*23*), pomegranate (*24*), and strawberry (*25*) juices and concentrates. The effect of sonication on anthocyanins in fruit juices has not been reported. The objective of this study was to investigate and model the effects of sonication on anthocyanin, ascorbic acid, and color of strawberry juice.

MATERIALS AND METHODS

Preparation of Strawberry Juice Samples. Fresh strawberries were purchased from a local fruit market (Begley's Marketing Services Ltd., Dublin, Ireland) and were frozen at −25 °C. Frozen strawberries were thawed overnight (12 h) at 4 °C and crushed using a domestic juice extractor (Kenstar, Dublin, Ireland). The obtained fresh juice was

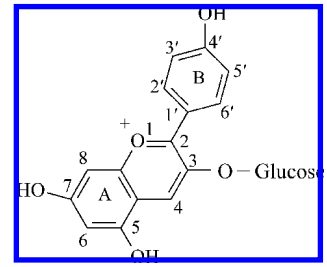

Figure 1. Structure of P3G.

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immediately filtered on a double layer cheese cloth to remove seeds and pulp from the juice and sonicated. Crushing of berries, juice extraction, and filtration were performed in a cold room maintained at 3 ± 1 °C.

Ultrasound Treatment. A 1500 W ultrasonic processor (VC 1500, Sonics and Materials Inc., Newtown, United States) with a 19 mm probe was used for sonication. Samples were processed at a constant frequency of 20 kHz. The energy input was controlled by setting the amplitude of the sonicator probe. Extrinsic parameters of amplitude (40, 50, 70, 90, and 100%) and treatment time (0, 3, 5, 7, and 10 min) were varied with pulse durations of 5 s on and 5 s off. The 100% amplitude corresponds to an amplitude of 61 μm. The acoustic energy densities dissipated in the juice sample were calculated as 0.33, 0.36, 0.47, 0.61, and 0.81 W/mL for amplitudes of 40, 50, 70, 90, and 100%, respectively, based on the formula presented by Tiwari et al. (*20*). Strawberry juice samples of 80 mL were placed in a 100 mL jacketed vessel through which water at 25 ± 1.0 °C with a flow rate of 0.5 L/min was circulated. At the 10 min treatment time, the maximum sample temperatures reached were 30.6, 31.6, 34.1, 37.2, and 39.9 °C for amplitudes levels of 40, 50, 70, 90, and 100%, respectively. The ultrasound probe was submerged to a depth of 25 mm in the sample. These experimental conditions were based upon a 5 log reduction of *E. coli* ATCC 25922 at an initial concentration of 1×10^6 CFU/mL. All treatments were carried out in triplicate.

Color Determination. Juice color was measured using a HunterLab colorimeter (ColorFlex, model A60-1010-615, Hunter Associates Laboratory Inc., Reston, VA). The instrument (65°/0° geometry, D25 optical sensor, 10° observer) was calibrated using white ($L = 92.8$; $a = -0.8$, $b = 0.1$) and black reference tiles. The color values were expressed as L^* (whiteness or brightness/darkness), a^* (redness/greenness), and b^* (yellowness/ blueness). The total color difference (TCD) (*26*) (eq 1) indicates the magnitude of color change after treatment (*27*). Color measurements were taken in triplicate from each sample with the mean values reported.

$$\mathrm{TCD} = \sqrt{(L^* - L_o)^2 + (a^* - a_o)^2 + (b^* - b_o)^2} \quad (1)$$

where L_o, a_o, and b_o are color values of untreated juice.

Anthocyanin Content. The anthocyanin content was determined following the high-performance liquid chromatography (HPLC) (Shimadzu model no. SPD-M10AVP, Shimadzu Co., Japan) validation and analytical procedure outlined by Zabetakis et al. (*28*), which involves extraction of 5 mL of sample with the mixture of methanol, acetic acid, and water in ratios of 25:1:24. Samples were centrifuged (Sanyo MSE Mistral 3000i, Sanyo, United Kingdom) for 10 min at 2000*g* and 4 °C. Five milliliters of the supernatant was filtered through 0.45 μm diameter PTFE syringe filters (Phenomenex, United Kingdom) and placed in an autosampler vial. The mobile phase was a solution comprising mixture of acetonitrile (83 mL), methanol (33 mL), and acetic acid (170 mL), which were mixed with trichloroacetic acid (0.65 g) that was previously dispersed in water to make up 1 L with distilled water. Separations were conducted on a Zorbax SB C_{18}, 5 μm, 150 mm × 4.6 mm column (Agilent Technologies, Dublin, Ireland). The sample loop was 20 μL with an isocratic flow rate of 1 mL/min, and the total run time was less than 8 min. Detection was carried out at 520 nm. For quantification, external calibration curves for P3G were prepared at concentrations from 25 to 100 μg/mL. Results are expressed as mean values.

Ascorbic Acid Determination. The ascorbic acid content was determined following the HPLC (Shimadzu model no. SPD-M10AVP, Shimadzu Co.) analytical procedure outlined by Lee and Coates (*29*). Ten microliter aliquot of samples were injected onto a Shimadzu C18 (15 cm × 4.6 cm, pore size 5 μm) coupled with HyperODS guard column. Twenty-five milliliter juice samples were pipetted into 50 mL centrifuge tubes containing 5 mL of 2.5% metaphosphoric acid. Samples were centrifuged for 10 min at 2000*g* and 4 °C (Sanyo MSE Mistral 3000ii, Sanyo). Five milliliters of the supernatant was filtered through 0.45 μm diameter PTFE syringe filters (Phenomenex) and placed in an autosampler vial. The mobile phase was 25 mM KH_2PO_4 (adjusted to pH 3.0 with phosphoric acid) with a flow rate of 1 mL/min. Eluate was monitored by UV detection at 245 nm. Chromatograms were recorded and processed with EZStart Chromatography Software V.7.2.1. Results were reported as mg/100 mL of strawberry juice.

Table 1. Independent Factors and Their Coded Levels Chosen for Central Composite Design

		coded level				
independent factor	symbol	$-\alpha$	-1	0	$+1$	$+\alpha$
amplitude (%)	X_1	40	50	70	90	100
time (min)	X_2	0	3	5	7	10

Other Analysis. The pH of treated and untreated strawberry juice samples was measured using a digital pH meter (model 420A, Orion Bench top pH meter, Allometrics Inc., Seabrook, United States). Samples (10 mL) were placed in a 50 mL beaker with a magnetic stirrer. Soluble solids were measured using a refractometer (Abbe 60, Bellingham + Stanley Ltd., United Kingdom). The refractive index was recorded and converted to °Brix. Measurements were carried out at 20.0 ± 0.5 °C. For titratable acidity, a sample of 20 mL was placed into a 250 mL beaker, and 80 mL of distilled water was added. This solution was then titrated against standardized 0.1 N NaOH (Sigma-Aldrich, Dublin, Ireland) to the phenolphthalein end point (pH = 8.2 ± 0.1). The volume of NaOH was converted to g anhydrous citric acid per 100 mL of juice (*30*).

Experimental Design. The effects of the two extrinsic parameters, namely, amplitude (X_1, %) and treatment time (X_2, min) on P3G (mg/100 mL), ascorbic acid (mg/100 mL), lightness (L^*), yellowness (a^*), redness (b^*), and TCD were investigated using response surface methodology. The experimental design employed was a central composite design with two independent variables each at five levels using Minitab statistical software (Minitab V.15.0). The experimental order was randomized. The levels of independent variables, namely, amplitude level (X_1, %) and treatment time (X_2, min), were selected based on values obtained in preliminary experiments. The amplitude level (X_1) was varied between 40 and 100% and treatment time (X_2) between 0 and 10 min. The coded values of the independent variables were $-\alpha$, -1, 0, $+1$, and $+\alpha$. The actual values and the corresponding values of two independent variables, X_1 and X_2, are listed in **Table 1**.

Experimental data from the central composite design were analyzed using response surface regression (SAS V.9.1.3) (**Table 2**) and fitted to a second-order polynomial model (eq 2).

$$Y = \beta_0 + \sum_{i=1}^{2} \beta_i X_i + \sum_{i=1}^{2} \beta_{ii} X_i^2 + \sum_i \sum_{j=i+1} \beta_{ij} X_i X_j \quad (2)$$

where Y is the predicted response, β_0 is the constant coefficient, β_i is the linear coefficient, β_{ii} is the quadratic coefficient, and β_{ij} is the cross product coefficient. X_i and X_j are independent variables. Three-dimensional curves of the response surface were developed using Minitab (V.15.0) software while holding the variables constant in the second-order polynomial model. The validity of the model was determined by comparing the experimental and predicted values.

RESULTS AND DISCUSSION

Table 3 shows the characteristics of the strawberry juice samples prior to sonication. The anthocyanin contents of strawberry juice expressed as P3G and ascorbic acid contents of strawberry juice were 44.21 and 53.30 mg/100 mL of juice, respectively. The anthocyanin and ascorbic acid contents of strawberries are influenced by various genetic (cultivar), environmental, and agronomic factors (*31*). The anthocyanin content for strawberry juice is reported to vary from 14.8 to 41.8 mg/100 g (*32*), although values as high as 50.3 mg/100 g have also been reported (*33*). Total soluble solids (%), pH, and titratable acidity as citric acid were 9.61, 3.14, and 0.73 g/100 mL, respectively. P3G, ascorbic acid, L^*, a^*, b^*, and TCD values were influenced by the two factors investigated, that is,

Table 2. Experimental Design Matrix, Coded Values, and Responses Studied

						Hunter color values			
		amplitude (%)	time (min)	P3G[c](mg/100 mL)	ascorbic acid (mg/100 mL)	L^*	a^*	b^*	TCD
sample no.[a]	run order[b]	X_1	X_2	Y_1	Y_2	Y_3	Y_4	Y_5	Y_8
1	3	70 (0)	5 (0)	43.81	49.99	33.23	39.31	24.67	11.66
2	6	90 (+1)	7 (+1)	43.22	47.4	35.35	40	23.45	13.64
3	7	50 (−1)	7 (+1)	43.95	49.86	33.82	38.98	25.52	11.56
4	2	90 (+1)	3 (−1)	43.7	49.85	33.46	41.47	26.68	9.77
5	5	70 (0)	5 (0)	44.18	49.65	33.23	39.31	24.65	11.67
6	12	50 (−1)	3 (−1)	44.22	51.2	31.94	39.88	27.62	8.62
7	1	70 (0)	5 (0)	44.45	49.98	33.28	39.27	24.61	11.75
8	11	70 (0)	5 (0)	43.97	49.26	33.18	39.36	24.73	11.57
9	10	70 (0)	0 (−α)	44.21	53.44	26.42	43.55	33.13	0.00
10	9	100 (+α)	5 (0)	43.62	47.98	34.85	40.75	25.02	12.03
11	13	40 (−α)	5(0)	44.25	51.46	33.26	39.77	25.91	10.64
12	4	70 (0)	10 (+α)	43.13	48.2	33.98	38.36	24.12	12.86
13	8	70 (0)	5 (0)	44.21	49.62	33.23	39.32	24.66	11.65
14	3	70 (0)	5 (0)	44.08	49.75	33.23	39.31	24.67	11.66

[a] Nonrandomized. [b] Randomized. [c] P3G (mg/100 mL).

Table 3. Characteristics of Strawberry Juice Prior to Sonication

parameters	
total soluble solids (°Brix)	9.61 ± 0.30
pH	3.14 ± 0.05
titratable acidity[a] (g/100 mL)	0.73 ± 0.01
anthocyanin[b] (mg/100 mL)	44.21 ± 0.90
ascorbic acid (mg/100 mL)	53.44 ± 0.43
lightness (L^*)	26.42 ± 1.19
redness (a^*)	43.55 ± 1.33
yellowness (b^*)	33.13 ± 1.09

[a] As citric acid. [b] As P3G.

ultrasound amplitude level (%) and treatment time (min). **Figure 2a**,**b** shows the chromatograms for anthocyanin and ascorbic acid degradation at a treatment time of 10 min.

Model Prediction and Fitting. The experimental results obtained as a function of ultrasound amplitude level and treatment time are shown in **Table 2**. P3G (mg/100 mL), ascorbic acid content (mg/100 mL), lightness (L^*), redness (a^*), yellowness (b^*), and TCD values varied between 44.45 and 43.13, 53.44 and 47.40, 35.35 and 26.42, 43.55 and 38.36, 33.13 and 23.45, and 0 and 13.64, respectively, whereas for control samples lightness (L_o), yellowness (a_o), and redness (b_o) values were 26.42, 43.55, and 33.13, respectively. During sonication, a slight increase (<1.0%) in anthocyanin content was observed at lower amplitude levels (**Figure 4a**). However, at higher amplitude levels and treatment time (>5 min), the anthocyanin content was found to decrease, with a maximum decrease of 3.2% observed at the maximum treatment conditions. The observed increase in anthocyanin content at lower amplitude levels and treatment times may be due to the extraction of pigments from the suspended pulp. Weak ultrasonic irradiation was reported to promote an increase in the amount of phenolic compounds found in red wine (*34*). The ascorbic acid content decreased with amplitude level and treatment time, with a decrease of 11% observed at the maximum treatment condition.

Table 4 shows the analysis of variance of the regression parameters for the predicted response surface quadratic models. The models had high correlation coefficients ($R^2 > 0.96$) and low coefficients of variance (CV ≤ 1.50) for ascorbic acid, L^*, a^*, and b^*. Lower correlations were obtained for P3G ($R^2 = 0.82$), and slightly higher CV values (5.44) were obtained for TCD. The significance of experimental factors that affects the treatment process may be quantified from the model coefficients, multiple determinations, and probabilities that were generated

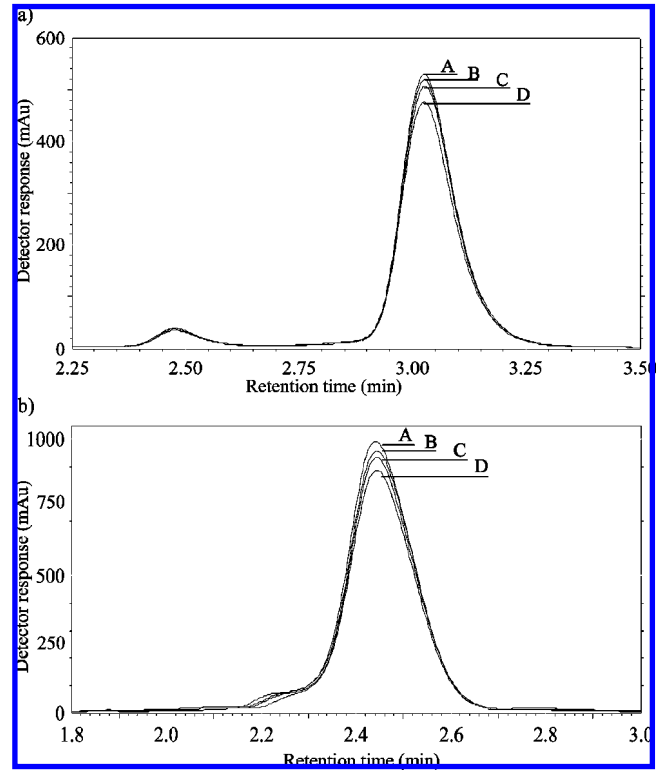

Figure 2. Chromatogram showing degradation of (**a**) anthocyanin and (**b**) ascorbic acid content (mg/100 mL) of strawberry juice during (A) control and sonicated at an amplitude level of (B) 40, (C) 70, and (D) 100% for a treatment time of 10 min.

from the RSREG procedure (SAS V.9.1.3). **Table 4** demonstrates that the analyses of variance (ANOVA) for the response surface quadratic model for various parameters were significant for ascorbic acid, L^*, a^*, b^*, TCD ($p < 0.0001$), and P3G ($p < 0.01$).

Canonical analysis of response surface models (**Table 5**) show that both linear and quadratic parameters were significant with $p < 0.0001$ (linear) for all of the parameters studied except for P3G ($p < 0.01$). The quadratic model was significant for L^*, b^*, TCD ($p < 0.0001$), a^* ($p < 0.001$), and ascorbic acid ($p < 0.01$) and insignificant for P3G. Both linear and quadratic models were significant for TCD ($p < 0.0001$) and ($p < 0.05$) for ascorbic acid, L^*, a^*, and b^*. The cross product for all of the models did not strongly fit the data except for ascorbic acid

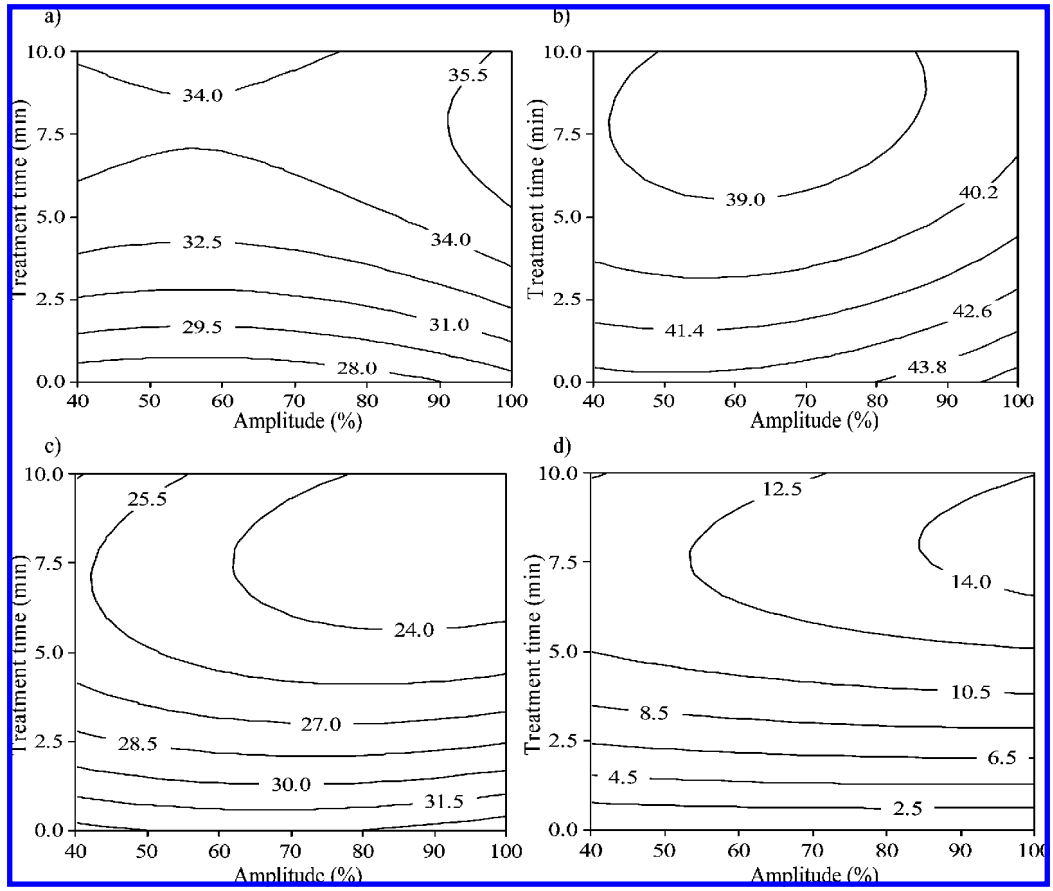

Figure 3. Contour plot illustrating the effect of amplitude (%) and treatment time (min) on (**a**) lightness (L^*), (**b**) redness (a^*), (**c**) yellowness (b^*), and (**d**) TCD.

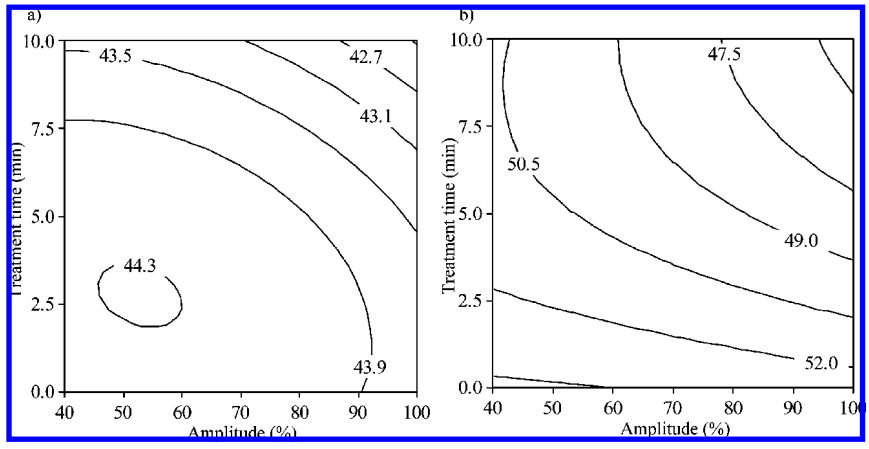

Figure 4. Contour plot illustrating the effect of amplitude (%) and treatment time (min) on (**a**) anthocyanin content (P3G, mg/100 mL) and (**b**) ascorbic acid content (mg/100 mL).

and TCD, which were both found to be significant ($p < 0.05$). The statistical analysis showed that the interaction among parameters was insignificant in every case. Thus, only linear and quadratic effects of the independent factors were the major determining conditions that influenced responses.

Table 6 shows the ANOVA of the independent factors obtained from RIDGE analysis of the responses studied. **Table 6** reveals that amplitude (X_1) significantly affected ascorbic acid ($p < 0.0001$), a^*, b^* ($p < 0.01$), and P3G ($P < 0.05$) and was insignificant for L^* and TCD. Treatment time (X_2) significantly affected ascorbic acid, L^*, a^*, b^*, TCD ($p < 0.0001$), and P3G ($p < 0.01$).

To confirm the adequacy of the fitted models, studentized residuals vs run order were tested, and the residuals were observed to be scattered randomly, suggesting that the variance of the original observations was constant for all responses (*35*). Furthermore, the normality assumption was satisfied as the residual plot approximated to a straight line for all six responses. To determine the accuracy of the predicted models, further experiments were carried out at constant treatment time of 10 min and varying amplitude level of 40, 50, 70, 90, and 100% under the same experimental conditions (**Figure 5a**,**b**). Predicted values for all of the parameters were found to be within the range of experimental values with coefficient of determinations (R^2) of 0.86, 0.99, 0.93, 0.72, and 0.95 for P3G, ascorbic acid, L^*, a^*, and b^*, respectively.

Analysis of Response Surfaces. The response surface model equations for the quality parameters are listed in eqs 3−8. The

Table 4. Regression Coefficients and ANOVA of Regression Parameters for the Predicted Response Surface Quadratic Models[a]

coefficient	P3G (Y_1)	ascorbic acid (Y_2)	L^* (Y_3)	a^* (Y_4)	b^* (Y_5)	TCD (Y_6)
β_0	43.18 (1.32)	53.98 (1.90)	30.315 (2.98)	45.71 (2.034)	36.879 (2.40)	−0.8578 (3.62)
			linear			
β_1	0.034^{ns} (0.03)	−0.0006^{ns} (0.04)	−0.130^{ns} (0.07)	−0.111* (0.045)	−0.124* (0.05)	0.0387^{ns} (0.08)
β_2	0.169^{ns} (0.20)	−0.466^{ns} (0.29)	1.887** (0.45)	−0.873* (0.30)	−1.934**** (0.36)	2.861*** (0.54)
			quadratic			
β_{11}	−0.0003^{ns} (0.00)	−0.0001^{ns} (0.00)	0.001** (0.00)	0.001** (0.00)	0.001* (0.00)	−0.00026^{ns} (0.00)
β_{22}	−0.0013* (0.003)	−0.007* (0.004)	0.0001**** (0.01)	−0.004*** (0.00)	−0.007**** (0.005)	0.00582**** (0.019)
			cross product			
β_{12}	−0.018^{ns} (0.007)	0.044* (0.010)	−0.120^{ns} (0.02)	0.066 (0.01)	0.158^{ns} (0.013)	−0.20886* (0.007)
R^2	0.82	0.97	0.97	0.96	0.98	0.98
CV	0.4832	0.6116	1.44	0.82	1.50	5.44
total model	0.0074	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001

[a] Parentheses indicate standard error. *, Significant at $p \leq 0.05$; **, significant at $p \leq 0.01$; ***, significant at $p \leq 0.001$; ****, significant at $p \leq 0.0001$; and ns, not significant.

Table 5. ANOVA of the Fitted Quadratic Models

	P3G (Y_1)		ascorbic acid (Y_2)		L^* (Y_3)		a^* (Y_4)		b^* (Y_5)		TCD (Y_6)	
regressions	F value	$p > F$	F value	$p > F$	F value	$p > F$	F value	$p > F$	F value	$p > F$	F value	$p > F$
linear	14.29	0.0023	144.72	<0.0001	75.73	<0.0001	76.43	<0.0001	166.6	<0.0001	141.76	<0.0001
quadratic	3.87	0.0669	11.17	0.0048	41.85	<0.0001	21.97	0.0006	79.73	<0.0001	63.69	<0.0001
cross product	0.24	0.6342	3.32	0.1061	0.00	0.9919	0.77	0.4072	2.16	0.1798	0.65	0.4451
total model	7.31	0.0074	63.02	<0.0001	47.03	<0.0001	39.51	<0.0001	98.96	<0.0001	82.31	<0.0001

Table 6. ANOVA of the Factors Obtained from RIDGE Analysis

	amplitude (X_1)		treatment time (X_2)	
independent variable	F value	$p > F$	F value	$p > F$
P3G (mg/100 mL)	5.1	0.0292	7.7	0.0096
ascorbic acid (mg/100 mL)	35.61	<0.0001	69.68	<0.0001
L^*	0.845	0.2817	340.82	<0.0001
a^*	11.41	0.0029	57.81	<0.0001
b^*	8.49	0.0072	159.89	<0.0001
TCD	3.61	0.0652	132.03	<0.0001

contour plots were made as a function of amplitude and treatment time. The effects of amplitude level and sonication time on quality parameters are illustrated in **Figures 3** and **4**. It can be seen that increased amplitude level and treatment time decrease P3G, ascorbic acid, a^* and b^* values and increase L^* values. However, there was a slight increase in P3G content at lower amplitude level and processing time (**Figure 4a**).

$$\text{P3G} = 43.18 - 0.034X_1 + 0.169X_2 - 0.0003X_1^2 - 0.0013X_2^2 - 0.018X_1X_2 \quad (3)$$

$$\text{AA} = 53.98 - 0.0006X_1 - 0.466X_2 - 0.0001X_1^2 - 0.007X_2^2 + 0.044X_1X_2 \quad (4)$$

$$L^* = 30.315 - 0.130X_1 + 1.887X_2 + 0.001X_1^2 + 0.0001X_2^2 - 0.120X_1X_2 \quad (5)$$

$$a^* = 45.71 - 0.111X_1 - 0.873X_2 + 0.001X_1^2 - 0.004X_2^2 - 0.066X_1X_2 \quad (6)$$

$$b^* = 36.879 - 0.24X_1 - 1.934X_2 + 0.001X_1^2 - 0.007X_2^2 + 0.158X_1X_2 \quad (7)$$

$$\text{TCD} = -0.8578 - 0.0387X_1 - 2.861X_2 - 0.00026X_1^2 + 0.00582X_2^2 - 0.20886X_1X_2 \quad (8)$$

Degradation Mechanisms. Cavitation involves the formation, growth, and rapid collapse of microscopic bubbles. Degradation of quality and nutritional parameters results from the extreme physical conditions that occur within the bubbles during cavitational collapse at microscale (*36*) and several sonochemical reactions occurring simultaneously or in isolation. The chemical effects produced by cavitation generate high local temperature (up to 5000 K), pressure (up to 500 MPa), and mechanical action

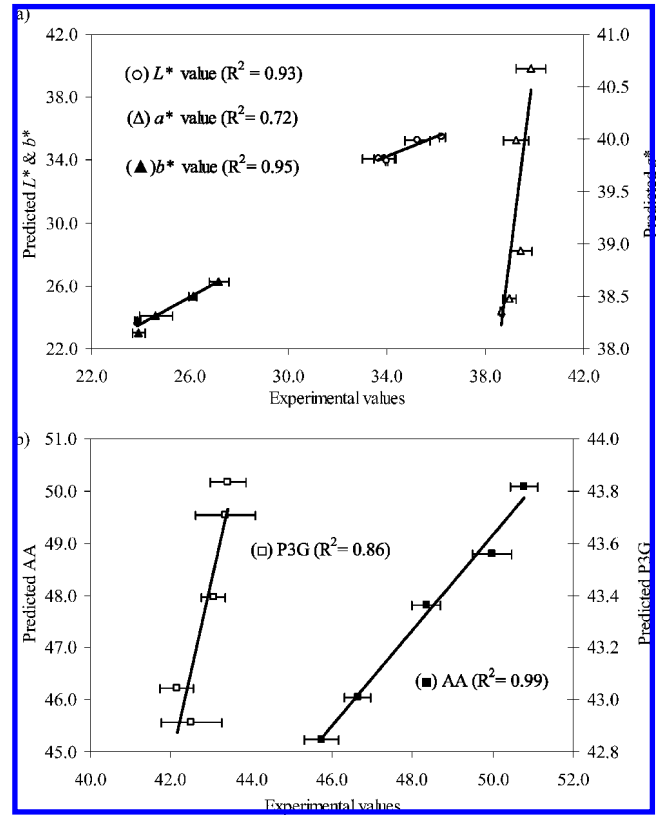

Figure 5. Correlation of predicted and experimental values for sonicated samples at 40, 50, 70, 90, and 100% amplitude levels for 10 min for (**a**) Hunter color values and (**b**) ascorbic acid (AA, mg/100 mL) and anthocyanin content (P3G, mg/100 mL).

between solid and liquid interfaces (*37, 38*). Zhao et al. (*39*) reported a similar mechanism for degradation of (all-*E*)-astaxanthin into unidentified colorless molecule(s) during extraction using sonication. Cavities formed by sonication may be filled with water vapor and gases dissolved in the juice, such as O_2 and N_2 (*40*). The anthocyanin and ascorbic acid degradation during ultrasonic processing could be related to oxidation reactions, promoted by the interaction of free radicals formed during sonication (*41*). Two reaction mechanisms have been proposed for sonodegradation. The first mechanism is pyrolysis within cavitation bubbles or nuclei; these are tiny, free-floating bubbles in the liquid or gas pockets trapped in the crevices of the solid boundaries in the liquid medium (*42*), which is likely to be the major reaction path for the degradation of polar compounds. Sonochemical reaction is the chemical manifestation of cavitation phenomena, and degassing of the medium enhances the yield of such a reaction and influences the phenomenon of radical formation by cavitation bubbles, which is the primary mechanism of a sonochemical reaction (*42*). The generation of $^{\bullet}OH$ radicals ($H_2O \rightarrow {}^{\bullet}OH + {}^{\bullet}H$) subsequently oxidizes the polar organic compounds.

The degradation pathway of ascorbic acid or anthocyanins could be due to either thermolysis and combustion occurring inside the bubble or reaction with hydroxyl radicals leading to the formation of oxidation products occurring on the bubble surface (*43*−*45*). Greater degradation of ascorbic acid as compared to anthocyanin could be due to differences in physicochemical properties of these compounds. Petrier et al. (*46*) reported that when two compounds with differing physicochemical properties are subjected to sonication, the more volatile compounds degrades first (*47*).

The level of anthocyanin degradation due to sonication is relatively low and compares favorably to thermal processing. Ultrasound processing technology may be considered as a potential alternative technique for juice processing where retention of nutritional quality is a priority.

LITERATURE CITED

(1) Tulipani, S.; Mezzetti, B.; Capocasa, F.; Bompadre, S.; Beekwilder, J.; Ric De Vos, C. H.; Capanoglu, E.; Bovy, A.; Battino, M. Antioxidants, phenolic compounds, and nutritional quality of different strawberry genotypes. *J. Agric. Food Chem.* **2008**, *56*, 696–704.

(2) Bailey, L. B.; Gregory, J. F. Folate metabolism and requirements. *J. Nutr.* **1999**, *129*, 779–782.

(3) Garzon, G. A.; Wrolstad, R. E. Comparison of the stability of pelargonidin-based anthocyanins in strawberry juice and concentrate. *J. Food Sci.* **2002**, *67*, 1288–1299.

(4) Wang, H.; Cao, G.; Prior, R. L. Oxygen radical absorbing capacity of anthocyanins. *J. Agric. Food Chem.* **1997**, *45*, 304–309.

(5) Duthie, G. G.; Duthie, S. J.; Kyle, J. A. M. Plant polyphenols in cancer and heart disease: Implications as nutritional antioxidants. *Nutr. Res. Rev.* **2000**, *13*, 79–106.

(6) Jackman, R. L.; Yada, R. Y.; Tung, M. A.; Speers, R. A. Anthocyanins as food colorants—A review. *J. Food Biochem.* **1987**, *11*, 201–247.

(7) Wang, W.; Xu, S. Degradation kinetics of anthocyanins in blackberry juice and concentrate. *J. Food Eng.* **2007**, *82*, 271–275.

(8) Shrikhan, A. J.; Francis, F. J. Effect of flavonols on ascorbic-acid and anthocyanin stability in model systems. *J. Food Sci.* **1974**, *39*, 904–906.

(9) Skrede, G.; Wrolstad, R. E.; Lea, P.; Enersen, G. Color stability of strawberry and blackcurrant syrups. *J. Food Sci.* **1992**, *57*, 172–177.

(10) Markakis, P.; Livingston, G. E.; Fellers, C. R. Quantitative aspects of strawberry pigment degradation. *Food Res.* **1957**, *22*, 117–130.

(11) Sondheimer, E.; Kertesz, Z. I. Participation of ascorbic acid in the destruction of anthocyanin in strawberry juice and model systems. *Food Res.* **1953**, *18*, 475–479.

(12) Mason, T. J.; Riera, E.; Vercet, A.; Lopez-Bueza, P. Application of ultrasound. In *Emerging Technologies for Food Processing*; Sun, D.-W., Ed.; Elsevier Ltd.: Cambridge, MA, 2005; Vol. 32, pp 3−351.

(13) Mason, T. J.; Paniwnyk, L.; Lorimer, J. P. The uses of ultrasound in food technology. *Ultrason. Sonochem.* **1996**, *3*, 253–260.

(14) Valero, M.; Recrosio, N.; Saura, D.; Munoz, N.; Martıc, N.; Lizama, V. Effects of ultrasonic treatments in orange juice processing. *J. Food Eng.* **2007**, *80*, 509–516.

(15) Dennis, J. D.; Todd, M. S.; Wu, J.; Mingruo, G. Inactivation of microorganisms in milk and apple cider treated with ultrasound. *J. Food Prot.* **2006**, *69*, 556–563.

(16) Cheng, L. H.; Soh, C. Y.; Liew, S. C.; Teh, F. F. Effects of sonication and carbonation on guava juice quality. *Food Chem.* **2007**, *104*, 1396–1401.

(17) Rui, M. S.; Cruz Vieira, M. C.; Silva, C. L. M. Modelling kinetics of watercress (*Nasturtium officinale*) colour changes due to heat and thermosonication treatments. *Innovative Food Sci. Emerging Technol.* **2007**, *8*, 244–252.

(18) Salleh-Mack, S. Z.; Roberts, J. S. Ultrasound pasteurization: The effects of temperature, soluble solids, organic acids and pH on the inactivation of *Escherichia coli* ATCC 25922. *Ultrason. Sonochem.* **2007**, *14*, 323–329.

(19) Knorr, D.; Zenker, M.; Heinz, V.; Lee, D. U. Applications and potential of ultrasonics in food processing. *Trends Food Sci. Technol.* **2004**, *15*, 261–266.

(20) Tiwari, B. K.; Muthukumarappan, K.; O'Donnell, C. P.; Cullen, P. J. Effects of sonication on the kinetics of orange juice quality parameters. *J. Agric. Food Chem.* **2008**, *56*, 2423–2428.

(21) Wang, W.; Xu, S. Degradation kinetics of anthocyanins in blackberry juice and concentrate. *J. Food Eng.* **2007**, *82*, 271–275.

(22) Cemeroglu, B.; Velioglu, S.; Isik, S. Degradation kinetics of anthocyanins in sour cherry juice and concentrate. *J. Food Sci.* **1994**, *59*, 1216–1218.

(23) Ochoa, M. R.; Kesseler, A. G.; Vullioud, M. B.; Lozano, J. E. Physical and chemical characteristics of raspberry pulp: Storage effect on composition and color. *Lebensm.-Wiss. Technol.* **1992**, *32*, 149–153.

(24) Martí, N.; Pérez-Vicente, A.; García-Viguera, C. Influence of storage temperature and ascorbic acid addition on pomegranate juice. *J. Sci. Food Agric.* **2001**, *82*, 217–222.

(25) Garcia-Viguera, C.; Bridle, P. Influence of structure on colour stability of anthocyanins flavylium salts with ascorbic acid. *Food Chem.* **1999**, *64*, 21–26.

(26) Tiwari, B. K.; Muthukumarappan, K.; O'Donnell, C. P.; Cullen, P. J. Modelling colour degradation of orange juice by ozone treatment using response surface methodology. *J. Food Eng.* **2008**, *88*, 553–560.

(27) Minolta. Precise Colour Communication: Colour Control from Feeling to Instrumentation; Minolta Report; Minolta: Japan, 1994.

(28) Zabetakis, I.; Leclerc, D.; Kajda, P. The effect of high hydrostatic pressure on the strawberry anthocyanins. *J. Agric. Food Chem.* **2000**, *48*, 2749–2754.

(29) Lee, H. S.; Coates, G. A. Vitamin C in frozen, fresh squeezed, unpasteurized, polyethylene-bottled orange juice: A storage study. *Food Chem.* **1999**, *65*, 165–168.

(30) Redd, J. B.; Hendrix, C. M.; Hendrix, D. L. *Quality Control Manual for Citrus Processing Plants, Book 1*; Intercity, Inc.: Safety Harbor, FL, 1986.

(31) Olsson, M. E.; Ekvall, J.; Gustavsson, K.; Nilsson, J.; Pillai, D.; Sjoholm, I.; Svensson, U.; Akesson, B.; Nyman, M. G. L. Antioxidants, low molecular weight carbohydrates, and total antioxidant capacity in strawberry (*Fragaria* × *ananassa*): Effects of cultivar, ripening, and storage. *J. Agric. Food Chem.* **2004**, *52*, 2490–2498.

(32) Wrolstad, R. E.; Putnam, T. P.; Varseveld, G. W. Color quality of frozen strawberries: Effect of anthocyanin, pH, total acidity, and ascorbic acid availability. *J. Food Sci.* **1970**, *35*, 448–452.
(33) Pilando, L. S.; Wrolstad, R. E.; Heatherbell, D. A. Influence of fruit composition, maturity and mold contamination on the color and appearance of strawberry wine. *J. Food Sci.* **1985**, *50*, 1121–1125.
(34) Masuzawa, N.; Ohdaira, E.; Ide, M. Effects of ultrasonic irradiation on phenolic compounds in wine. *Jpn. J. Appl. Phys.* **2000**, *39*, 2978–2979.
(35) Myers, R. H.; Montgomery, D. C. *Response Surface Methodology: Process and Product Optimization Using Designed Experiments*, 2nd ed.; Wiley: New York, 2002.
(36) Suslick, K. S. *Ultrasounds: Its Chemical, Physical and Biological Effects*; VHC Publishers: New York, 1988.
(37) Contamine, R. F.; Wilhelm, A. M.; Berlan, J.; Delmas, H. Power measurement in sonochemistry. *Ultrason. Sonochem.* **1995**, *2*, 43–47.
(38) Suslick, K. S.; Hammerton, D. A.; Cline, R. E. Thesonoche medical hot spot. *J. Am. Chem. Soc.* **1986**, *108*, 5641–5642.
(39) Zhao, L.; Zhao, G.; Chen, F.; Wang, Z.; Wu, J.; Hu, X. Different effects of microwave and ultrasound on the stability of (all-*E*)-astaxanthin. *J. Agric. Food Chem.* **2006**, *54*, 8346–8351.
(40) Korn, M.; Prim, P. M.; deSousa, C. S. Influence of ultrasonic waves on phosphate determination by the molybdenum blue method. *Microchem. J.* **2002**, *73* (3), 273–277.
(41) Portenlänger, G.; Heusinger, H. Chemical reactions induced by ultrasound and γ-rays in aqueous solutions of L-ascorbic acid. *Carbohydr. Res.* **1992**, *232*, 291–301.
(42) Sivasankar, T.; Paunikar, A. W.; Moholkar, V. S. Mechanistic approach to enhancement of the yield of a sonochemical reaction. *AIChE J.* **2007**, *53*, 1132–1143.
(43) Hart, E. J.; Henglein, A. Free radical and free atom reactions in the sonolysis of aqueous iodide and formate solutions. *J. Phys. Chem.* **1985**, *89*, 4342–4347.
(44) Riesz, P.; Berdhal, D.; Christman, C. L. *Environm. Health Pespect.* **1985**, *64*, 233–252.
(45) von Sonntag, C.; Mark, G.; Tauber, A.; Schuchmann, H. P. *Advances in Sonochemistry*; JAI Press: London, 1999; Vol. 5, pp 109−132.
(46) Petrier, C.; Combet, E.; Mason, T. Oxygen-induced concurrent ultrasonic degradation of volatile and non-volatile aromatic compounds. *Ultrason. Sonochem.* **2007**, *14*, 117–121.
(47) Petrier, C.; Jiang, Y.; Lamy, M.-F. Ultrasound and environment: Sonochemical destruction of chloroaromatic derivatives. *Environ. Sci. Technol.* **1998**, *32*, 1316–1318.

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