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Kinetics of Freshly Squeezed Orange Juice Quality Changes during Ozone Processing

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Freshly squeezed orange juice samples were ozonated with control variables of gas flow rate (0−0.25 L min^{-1}), ozone concentration (0.6−10.0%w/w), and treatment time (0−10 min). Effects of ozone processing variables on orange juice quality parameters of pH, °Brix, titratable acidity (TA), cloud value, nonenzymatic browning (NEB), color values (L^*, a^*, and b^*), and ascorbic acid content were determined. No significant changes in pH, °Brix, TA, cloud value, and NEB ($p < 0.05$) were found. L^*, a^*, and b^* color values were significantly affected by gas flow rate, ozone concentration, and treatment time. The changes in lightness (L^*) values and total color difference (TCD) values were fitted well to zero-order kinetics, whereas a^*, b^*, and ascorbic acid degradation followed first-order kinetics. The rate constants for a^*, b^*, and TCD were linearly correlated with ozone concentration ($R^2 = 0.88-0.99$), whereas the rate constants for L^* and ascorbic acid were exponentially correlated ($R^2 = 0.94-0.98$).

KEYWORDS: Ozone; orange juice; color degradation; ascorbic acid degradation

INTRODUCTION

Recently, a number of commercial fruit juice processors in the United States and Europe have started to employ ozone for pasteurization of juices, resulting in industry guidelines being issued by the U.S. Food and Drug Administration (FDA) (*1*). However, these guidelines highlight gaps in the literature with respect to the effect of ozone on juice quality parameters. Ozone is an effective sanitizer with strong disinfecting properties. Ozone rapidly decomposes into oxygen, leaving no toxic residues, making it environmentally friendly (*2*). Ozone decomposes to produce numerous free radicals, predominantly hydroxyl free radicals, which increase at higher temperature and pH (*3*). The mechanism of ozone self-decomposition has been well documented (*4–6*). Such advantages make ozone processing an attractive option for the food industry. Ozone has been declared as generally recognized as safe (GRAS) for use in food processing by the FDA in 1997 (*3*).

Ozone has been studied for fruit juice processing including apple cider (*7, 8*). Williams et al. (*9*) studied the effect of ozone in combination with dimethyl dicarbonate and hydrogen peroxide for orange juice preservation. They reported that a 5 log reduction of *Escherichia coli* O157:H7 could be achieved using ozone in combination with dimethyl dicarbonate. A number of studies reported the effects of ozone on quality parameters of treated fruits and vegetables (*10, 11*).

The nutritional quality of orange juice is primarily related to the ascorbic acid content (*12*). Ascorbic acid is thermolable and highly sensitive to various processing and storage conditions. The main factors affecting ascorbic acid loss in orange juice include temperature, salt and sugar concentration, pH, oxygen, enzymes, light, metal catalysts, and initial ascorbic acid concentration (*13*). Ascorbic acid degradation reactions are often responsible for important quality changes that occur during the storage of foods, limiting shelf life (*14*). However, the effect of ozone on the color and ascorbic acid degradation of orange juice has not been reported. Ozonation of liquid phases is most frequently accomplished by injecting ozone gas (mixtures of air/ozone or oxygen/ozone) through a sparger into a liquid. Typically, studies on ozone absorption in the aqueous systems are carried out in stirred-tank reactors or bubble columns.

The FDA's approval of ozone as a direct additive to food has triggered interest in ozone applications among academic researchers and food processors (*15*). Given the extremely limited data available in the literature on the high reactivity and instability of ozone, it is difficult to predict its reaction in the presence of organic matter (*16*). It may oxidize or ionize a substrate or spontaneously decompose to oxygen and free radicals. Kinetic models can be used for an objective, fast, and economic assessment of food safety. Kinetic modeling may also be employed to predict the influence of processing on critical

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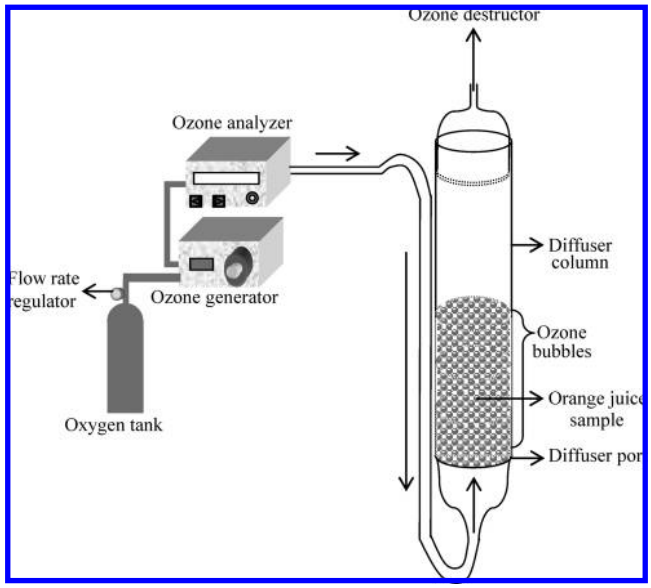

Figure 1. Schematic of ozone treatment system.

quality parameters. Kinetic models for microbial inactivation using ozone as a disinfecting agent are reported by various workers (*17, 18*). However, kinetics models of orange juice quality parameters during ozone processing are not reported in the literature. Hence, the objective of this study was to investigate the kinetics of freshly squeezed orange juice quality changes during ozone processing as a function of gas flow rate, ozone concentration, and treatment time.

MATERIALS AND METHODS

Preparation of Orange Juice Samples. Oranges (*Citrus sinensis* var. Valencia) harvested in July 2007 were purchased from a local supplier (Reilly Wholesale Ltd., Dublin, Ireland). Fresh juice was squeezed using a table-top citrus juice extractor (Braun Gmbh, Kronberg, Germany), yielding ca. 35% juice from 470 oranges, that is, 50−60 mL of juice per orange weighing approximately 150 ± 4.5 g. Juice pulp was removed by filtering with a double layer of cheesecloth.

Ozone Treatment. Experiments were carried out in a 250 mL bubble column with an built-in diffuser (**Figure 1**). Ozone was generated using an ozone generator (model OL80, Ozoneservices, Canada). Oxygen flow rate was controlled using a gas flow regulator. The experimental design for this work was based upon a parallel inactivation study for *E. coli* O157:H7, using the same control conditions. A 6 log reduction was achieved in under 6 min at an optimum flow rate of 0.125 L min^{-1} and a maximum ozone concentration obtainable (4.8%) at this flow rate. Ozone concentration in the gas supply was varied (0.6−10.0% w/w of oxygen) and recorded using an ozone gas analyzer (model OLA-DLS, Ozoneservices). Samples were ozonated at various gas flow rates (0.25, 0.125, 0.0625, and 0.0312 L min^{-1}). Ozone concentration was governed by the gas flow rate with the maximum concentration achievable at the lowest flow rates. Hence, ozone concentration ranges for these flow rates were 0.6−3.0, 1.0−4.8, 1.6−7.8, and 2.3−10.0% w/w, respectively. Samples were treated for 0 (control), 2, 4, 6, 8, and 10 min processing times. Treatments were performed at 20 ± 0.5 °C.

pH, Soluble Solids (°Brix), and Titratable Acidity. The pH of treated and untreated orange juice samples was measured using a digital pH-meter (Orion model 420A, Allometrics Inc., Seabrook, TX). Continually stirred samples (10 mL) were measured at 20 ± 0.5 °C. Soluble solids were measured using a refractometer (Abbe 60, Bellingham + Stanley Ltd., U.K.). Refractive index was recorded and converted to °Brix. Measurements were performed at 20 ± 0.5 °C. The refractometer prism was cleaned with distilled water after each analysis.

To determine titratable acidity (TA), 20 mL samples were diluted with 80 mL of distilled water and 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 grams of citric acid per 100 mL of juice, and TA was calculated using eq 1

$$\mathrm{TA} = \frac{[V \times 0.1\ \mathrm{N\ NaOH} \times 0.067 \times 100]}{m} \quad (1)$$

where V is volume (mL) of NaOH and m is mass of orange juice (g).

Color, Nonenzymatic Browning, and Cloud Value. Orange juice color was measured using a HunterLab colorimeter (ColorFlex modelA60-1010-615, Hunter Associates 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). Total color difference (TCD) was determined using eq 2, which indicates the magnitude of color change after treatment. Color measurements were taken in triplicate.

$$\mathrm{TCD} = \sqrt{(L^* - L_0)^2 + (a^* - a_0)^2 + (b^* - b_0)^2} \quad (2)$$

where L_0, a_0, and b_0 are the color values of control juice samples.

Nonenzymatic browning was measured using the method developed by Meydav et al. (*19*). Ten milliliter orange juice samples were centrifuged (10 min, 756*g*) (Sigma 1A, AGB Scientific Ltd., Dublin, Ireland) at 20 °C to remove coarse particles. Five milliliters of ethyl alcohol (95%, Sigma-Aldrich, Dublin, Ireland) was added to 5 mL of juice supernatant and centrifuged as above. The absorbance of the supernatant was obtained at 420 nm using a Unicam UV−vis (UV2) spectrophotometer.

Cloud value of orange juice was determined as outlined by Versteeg et al. (*20*). Five milliliter orange juice samples were centrifuged as outlined above. Cloud value was determined as the supernatant absorbance at 660 nm using a Unicam UV−vis (UV2) spectrophotometer with distilled water serving as a blank.

Ascorbic Acid Determination. Ascorbic acid content was determined following the HPLC (Simadzu model SPD-M10AVP) analytical procedure outlined by Lee and Coates (*21*). Ten microliter aliquot

Table 1. Effect of Ozonation on the Quality Parameters at a Treatment Time of 10 min[a]

		gas flow rate and ozone concentration			
parameter	control	0.03 L min^{-1} 10.0% w/w	0.06 L min^{-1} 7.8% w/w	0.125 L min^{-1} 4.8% w/w	0.25 L min^{-1} 3.0% w/w
pH	3.43 ± 0.04a	3.42 ± 0.02a	3.43 ± 0.01a	3.40 ± 0.06a	3.44 ± 0.02a
°Brix	9.90 ± 0.42a	10.09 ± 0.47a	10.09 ± 0.47a	9.90 ± 0.42a	9.90 ± 0.42a
TA	0.67 ± 0.02a	0.67 ± 0.01a	0.67 ± 0.00a	0.68 ± 0.00a	0.68 ± 0.00a
cloud	0.426 ± 0.013a	0.427 ± 0.00a	0.426 ± 0.01a	0.428 ± 0.04a	0.432 ± 0.02a
NEB	0.021 ± 0.01a	0.020 ± 0.02a	0.021 ± 0.00a	0.020 ± 0.01a	0.020 ± 0.01a
L^*	58.42 ± 0.59a	59.90 ± 0.17b	60.16 ± 0.67c	62.06 ± 0.67d	62.46 ± 0.65e
a^*	10.45 ± 0.61a	6.82 ± 1.01b	5.95 ± 0.68c	2.52 ± 1.32d	1.460 ± 0.96e
b^*	59.65 ± 0.23a	52.94 ± 0.05b	48.80 ± 0.38c	40.95 ± 0.20d	34.93 ± 0.25e
ascorbic acid	41.59 ± 0.18a		12.70 ± 0.31b		

[a] Values followed by same letter in a row are not significantly different ($p < 0.05$).

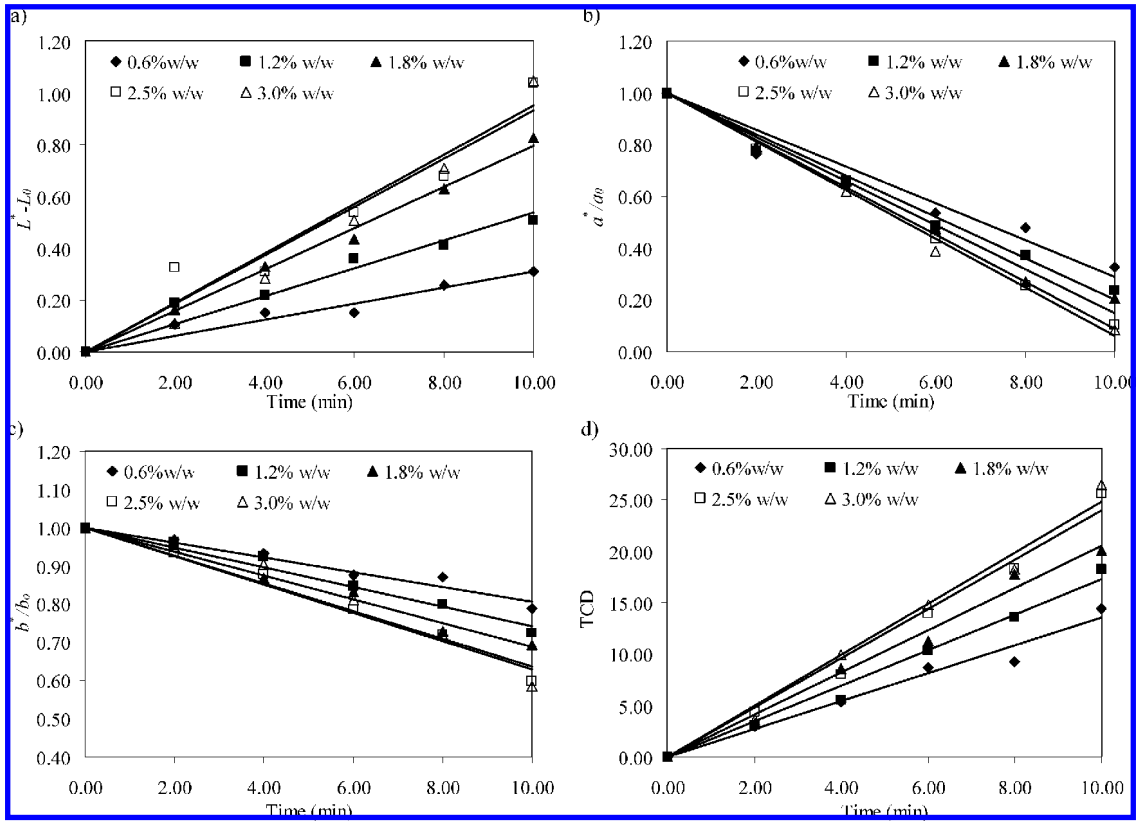

Figure 2. Changes in (**a**) lightness value ($L^* - L_0$), (**b**) red-green value (a^*/a_0), (**c**) blue-green value (b^*/b_0), and (**d**) total color difference (TCD) of orange juice during ozonation at a gas flow rate of 0.25 L min^{-1} with various concentrations.

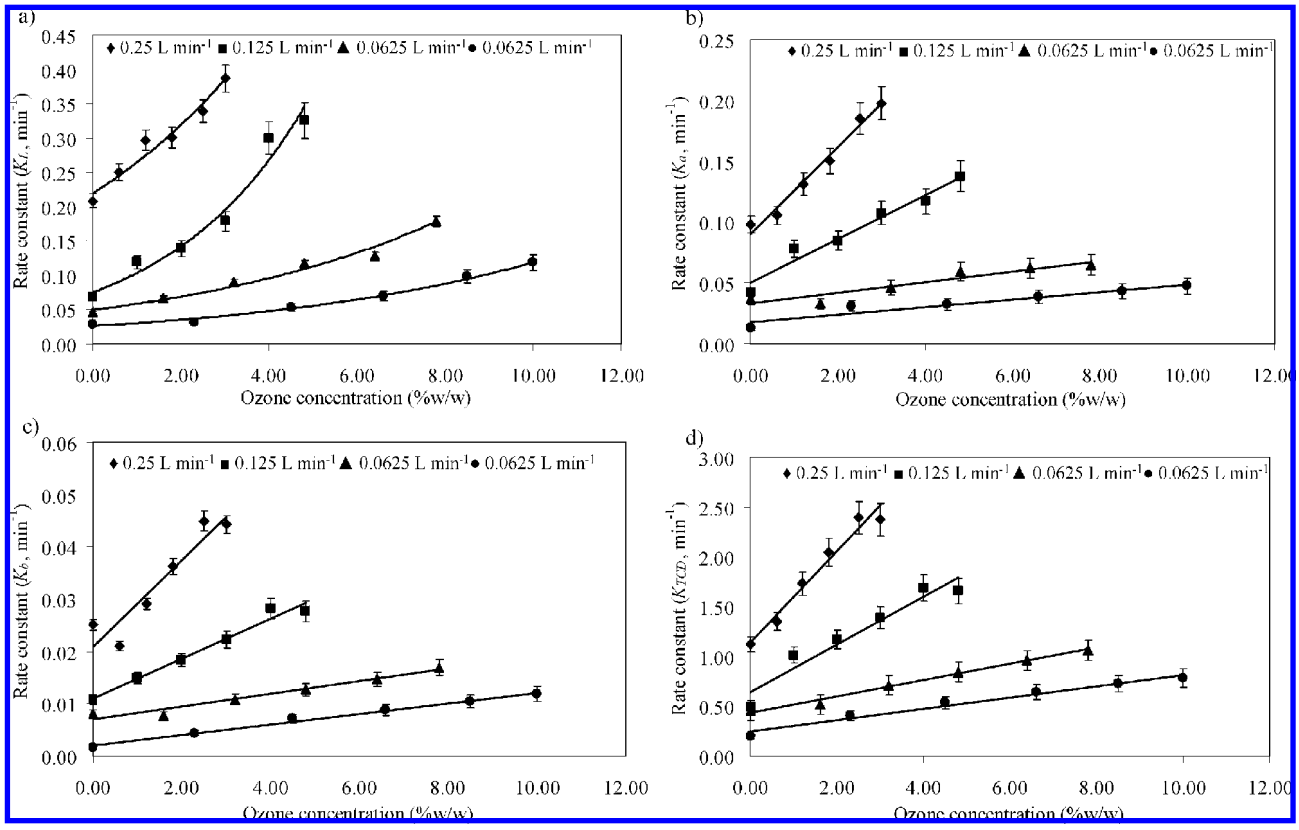

Figure 3. Effect of ozone concentration (% w/w) on reaction rate constants for (**a**) L^* value (K_L, min^{-1}), (**b**) a^* value (K_a, min^{-1}), (**c**) b^* value (K_b, min^{-1}), and (**d**) TCD value (K_{TCD}, min^{-1}) of orange juice during ozonation at gas flow rates of (◇) 0.25 L min^{-1}, (□) 0.125 L min^{-1}, (△) 0.0625 L min^{-1}, and (○) 0.0312 L min^{-1}, respectively.

samples were injected into a Shimadzu C18 (15 cm × 4.6 cm, pore size = 5 μm) coupled with a 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 centri-

Table 2. Regression Equation for the Rate Constants (y, min^{-1}) versus Ozone Concentration (x, % w/w) at Different Gas Flow Rates

gas flow rate,	L^*			a^*			b^*			TCD		
L min^{-1}	eq	SE[a]	R^2	eq	SE	R^2	eq	SE	R^2	eq	SE	R^2
0.250	$y = 0.2191e^{0.1879x}$	0.061	0.96	$y = 0.0357x + 0.091$	0.040	0.98	$y = 0.0083x + 0.021$	0.010	0.88	$y = 0.4569x + 1.145$	0.51	0.97
0.125	$y = 0.0752e^{0.3179x}$	0.058	0.97	$y = 0.0181x + 0.050$	0.018	0.96	$y = 0.0038x + 0.011$	0.004	0.97	$y = 0.2385x + 0.646$	0.26	0.93
0.0625	$y = 0.0497e^{0.1641x}$	0.016	0.98	$y = 0.0044x + 0.033$	0.006	0.89	$y = 0.0012x + 0.007$	0.001	0.96	$y = 0.0822x + 0.432$	0.07	0.99
0.0312	$y = 0.0261e^{0.1515x}$	0.022	0.98	$y = 0.0031x + 0.018$	0.008	0.92	$y = 0.0010x + 0.002$	0.002	0.99	$y = 0.0572x + 0.243$	0.13	0.98

[a] Standard error.

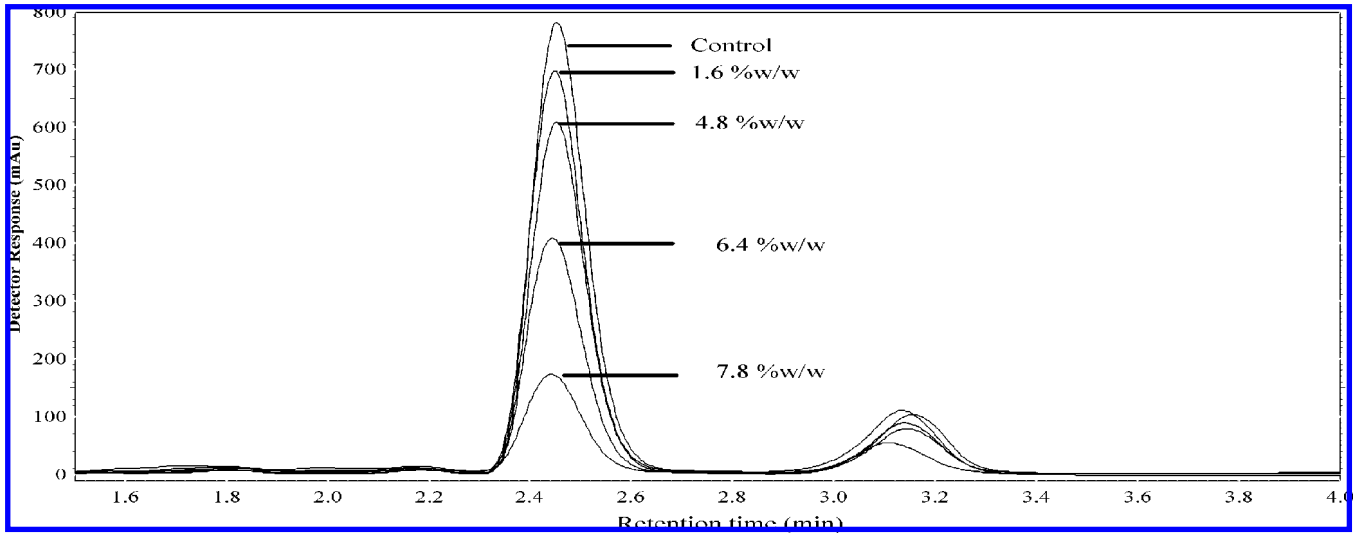

Figure 4. Chromatogram showing degradation of orange juice ascorbic acid (mg/100 mL) during ozone treatment.

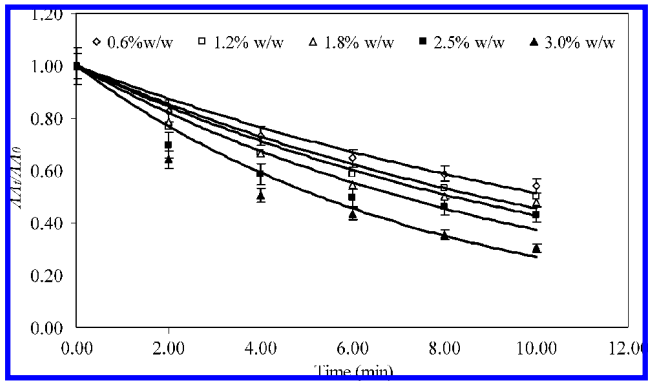

Figure 5. Changes in ascorbic acid content (mg/100 mL of orange juice) with treatment time (min) at constant flow rate of 0.0625 L min^{-1} and ozone concentration.

fuged (Sanyo MSE Mistral 3000i) for 10 min at 2000g and 4 °C. Five milliliters of the supernatant was filtered through PTFE syringe filters (0.45 μm, 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. Quantification was performed by comparing the chromatographic peak area with that of the external standard.

Twenty-five milligrams of standard ascorbic acid (Sigma Aldrich) was dissolved in 50 mL of mobile phase to obtain the calibration curve, a plot in the concentration range of 0.5−100 mg/L and based on a 10-point calibration. The calibration graph was linear over the range ($R^2 = 0.998$). Standard solutions and sample extracts were filtered through a prefilter and then a 0.45 μm Millipore membrane before their injection. To prevent the loss of ascorbic acid, standard solutions and extracted orange juice samples were protected from light covered with aluminum foil using amber flasks. The limits of detection (LOD) and quantification (LOQ) were determined by serial dilutions of ascorbic acid solutions to obtain signal/noise ratios (S/N) of 3 for LOD and 10 for LOQ. Chromatograms were recorded and processed with EZStart Chromatography software V.7.2. 1. Results were reported as milligrams per 100 mL of orange juice.

Kinetics Analysis. Kinetic models were developed using a two-step procedure (*22*). Reaction rate constants were determined by fitting the experimental data to zero-order (eq 3) and first-order (eq 4) kinetic models

$$C = C_0 + k_0 t \quad (3)$$

$$C = C_0 \, e^{k_1 t} \quad (4)$$

where C is the studied parameter (L^*, a^*, b^*, TCD) at any given reaction time, C_0 is the initial value of an untreated samples (L_0, a_0, b_0), and k_0 and k_1 are rate constants.

In the second step the rate constants were modeled as a function of ozone concentration. Data fitting was considered to be significant at a probability level of 95%. Statistical analysis and model parameters of the kinetic models (k_0 and k_1) were estimated using the PROC NLIN program of a nonlinear regression (SAS version 9.1, SAS Institute, Cary, NC).

Experimental Design and Statistical Analysis. A general factorial design (SAS V.9.1, SAS Institute) consisting of 240 experimental trials was employed. During ozone treatment, the effects of gas flow rate (L min^{-1}), ozone concentration (% w/w), and treatment time (min) were studied. Analysis of variance (ANOVA) was carried out to determine any significant differences ($p < 0.05$) among the applied treatments.

RESULTS AND DISCUSSION

pH, °Brix, and Titratable Acidity. The mean values obtained for pH, °Brix, and titratable acidity of control and treated orange juices samples at different gas flow rates are shown in **Table 1**. No significant changes in pH, °Brix, and titratable acidity of ozonated samples were observed. Comparatively, Ho (*23*) reported no significant differences in the pH, °Brix, and titratable acidity values of orange juice treated with dense phase carbon dioxide. Similarly, no significant changes

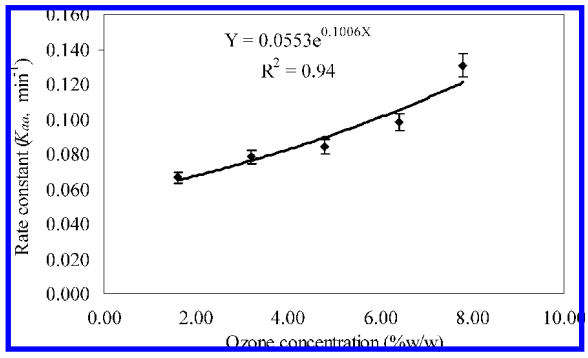

Figure 6. Change in ascorbic acid rate constant (min^{-1}) with respect to ozone concentration (% w/w).

were observed by Yagiz et al. (*24*) for mandarin juice processed by dense phase carbon dioxide. The literature suggests that nonthermal food processes have generally no significant effect on pH, °Brix, and titratable acidity values as was observed in the case of ultrasonic processing of orange juice (*25, 26*) and apple cider (*27*), ozone processing of apple cider (*7*), and pulse electric field processing of orange juice (*28*). However, during a storage period of 21 days a significant decrease in the °Brix of ozonated apple cider was reported by Choi and Nielsen (*7*).

Cloud Value and Nonenzymatic Browning. No significant differences ($p < 0.05$) in browning and cloud value of ozonated samples were found (**Table 1**). Choi and Nielsen (*7*) also reported no significant change in cloud value in apple cider after ozone treatment, but they observed higher levels of sedimentation after 21 days of storage. A similar trend was observed in this study (data not reported). Fruit juice cloud is mainly caused by suspended particles in colloidal form maintained by pectin molecules. Cloud loss or clarification occurs mainly due to the enzymatic activity of pectinesterase (PE), which de-esterifies pectin. PE attacks a methyl ester group adjacent to a free carboxyl group and cleaves these methyl esters, producing methanol and polygalacturonic acid (*29, 30*). As consumers associate cloud loss with spoilage and quality degradation, no reduction in cloud value is desirable.

Browning is a common problem encountered during juice processing, which results from the action of a group of enzymes called polyphenol oxidases, often referred to as enzymatic browning. Nonenzymatic browning may result from the condensation of a carbonyl group with amino acids (Maillard reaction). However, sugars and ascorbic acid also undergo browning reactions in the absence of free amino acids (caramelization), and many of the products formed are similar to those resulting from the Maillard reaction (*31*) and also due to particulate fractions (*32*). Ozone is a strong oxidizing agent, and browning is an oxidative reaction process (*33*) caused by the oxidation of phenols (*34*). However, ozone has been reported to prevent browning reactions in fresh-cut potatoes (*35*). No significant browning was observed in this study.

Color Kinetics. Ozonated samples were observed to be lighter in color, that is, increased L^* value, whereas a^* and b^* values of orange juice samples were found to decrease. These changes were a function of ozone concentration, gas flow rate, and treatment time. Mean L^* value increased from 58.42 to 62.46, whereas mean a^* and b^* values decreased from 10.45 to 1.46 and from 59.65 to 34.93, respectively, with an increase in gas flow rate from 0.0312 to 0.25 L min^{-1} (**Table 1**).

Relative changes in lightness (L^*), red-green (a^*), blue-yellow (b^*) values, and TCD values with reference to the control as a function of treatment time (min) for various ozone concentration levels at a gas flow rate of 0.25 L min^{-1} are shown in **Figure 2**. A zero-order kinetics model fitted well to ($p < 0.05$) $L^* - L_0$ and TCD values, whereas first-order models fitted well to a^*/a_0 and b^*/b_0 values with all coefficients of determination (R^2) greater than 0.94. Reaction rate constants (K_L, K_a, K_b, and K_{TCD}) were obtained using eqs 3 and 4. The reaction rate constants were evaluated for each gas flow rate as a function of ozone concentration using linear, exponential, and polynomial models. The effect of ozone concentration on the rate constants (K_L, K_a, K_b, K_{TCD}) for L^*, a^*, b^*, and TCD are shown in **Figure 3**.

Reaction rate constants (K_L) for L^* followed an exponential increase with respect to ozone concentration (% w/w) at each gas flow rate with high coefficient of determination values (0.96−0.98) and low standard errors (0.016−0.061) as shown in **Table 2**. For a^*, b^*, and TCD values, the reaction rate constants increased linearly with ozone concentration (% w/w) at each gas flow rate with high coefficient of determination values (0.88−0.99) and low standard errors (0.001−0.51) as shown in **Table 2**. **Figure 3** shows nonzero reaction rate constants (K_L, K_a, K_b, K_{TCD}) with no ozone in the feed gas, indicating degradation solely due to oxygen. These results demonstrate the significant effect of oxygen and ozone on the color degradation of orange juice samples. The oxygen proportion in the feed gas plays a synergistic role in color degradation.

Color degradation of fruit juices due to ozone processing has not been reported to date; however, comparisons may be made with other nonfood applications. The strong oxidizing potential of ozone is derived from the nascent oxygen atom (*2*). It has been reported that ozonation of organic dyes leads to color loss as a result of oxidative cleavage of chromophores (*36*) due to the breakdown of conjugated double bonds. Similarly, the chromophore with conjugated double bonds of carotenoid pigments may be degraded as above. Carotenoid pigments, which contribute to yellow, orange, or red color in orange juice, contain one or more aromatic rings (*37, 38*). The ozone and hydroxyl radicals (OH^-) generated in the aqueous solution may open these aromatic rings and lead to partial oxidation of products such as organic acids, aldehydes, and ketones.

Ascorbic Acid Degradation. The ascorbic acid content of ozonated samples was studied at a gas flow rate of 0.0625 L min^{-1}. The ascorbic acid content was found to decrease from 41.59 to 12.70 mg/100 mL after 10 min of treatment time (**Table 1**). **Figure 4** shows the chromatogram for ascorbic acid degradation at a constant flow rate of 0.0625 L min^{-1}. Ascorbic acid degradation was found to follow first-order kinetics ($R^2 > 0.92$, SE < 0.1) (**Figure 5**). A similar result was reported for other juices when exposed to different oxygen levels (*39–42*), which suggests aerobic degradation of ascorbic acid. The ascorbic acid degradation rate constant (K_{aa}) was found to increase exponentially with respect to ozone concentration (**Figure 6**, $R^2 = 0.94$, SE = 0.095). The degradation of ascorbic acid is known to occur by both oxidative and nonoxidative mechanisms (*43*). Molecular ozone reactions are selective and limited to unsaturated aromatic and aliphatic compounds. Ozone oxidizes organic matter via two pathways, by direct oxidation with ozone molecules (eq 5) and by the generation of free radical intermediates, such as OH^- (eq 6), a powerful, effective, nonselective oxidizing agent (*44, 45*).

$$O_3 + C \rightarrow C_{ox} \quad (5)$$

$$O_3 \xrightarrow{OH^-} OH \xrightarrow{C} C_{ox} \quad (6)$$

However, a study by Garcia-Viguera and Bridle (*46*) with model systems indicated that the degradation is more likely due to the free radical mechanism.

The effects of ozone concentration, treatment time, and gas flow rate on selected quality parameters of freshly squeezed orange juice were evaluated. No significant differences in pH, °Brix, TA, NEB, and cloud values were found. However, ozonation was found to have a significant effect on juice color and ascorbic acid content. It is also evident that the oxygen component of the feed gas has a synergistic effect on the degradation of these quality parameters. These findings should be considered prior to industrial adoption of this technology in fruit juice applications.

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