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Removal of phenolic, turbidity and color in sugarcane juice by electrocoagulation as a sulfur-free process

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Running Title: Removal of color in sugarcane juice by electrocoagulation

Abstract

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1. Introduction

Sugarcane juice is an important Brazilian feedstock for sugar, ethanol and bioenergy. Sugarcane is a complex material and its composition and characteristic are influenced by climate, phytosanitary, soil, harvest process, age and cultivars. Among the parameters measure on sugarcane juice, color stands out as the most important given the difficulty of its removal, leading to an increase in process cost of crystal sugar (Nguyen et al. 2015; Sartori et al., 2017a; Sartori et al., 2015b; Sartori et al., 2017c). Usually, the low color in crystal sugar means that its value will be high (Clarke and Godshall, 1988). There are four color-causing agents in cane sugar processes: plant pigments (chlorophylls, xanthophylls and anthocyanins), Maillard´s reaction, caramelization and alkaline fructose degradation products (Rein, 2007). In general, the sugarcane color is related to enzymatic reactions with phenolic compounds naturally in sugarcane (Nguyen and Doherty, 2012). Phenolic compounds have a hydroxyl group attached to an aromatic ring and are classified as: simple phenols or polyphenols, according to number of phenol unities. In the sugarcane juice, they are in a form of phenolic acids as hydroxycinamic acids (for example caffeic and sinapinic acids) and flavonoids (for example, vitexin and diosmetin-8-*C*-glycoside) (Abbas et al., 2014; Duarte-Almeida et al., 2006; Colombo et al., 2006), however, pigmentation was more related to polymerized phenolic compounds than simple phenolic compounds with low molecular weight (Clarke and Godshall, 1988).

In Brazilian sugarcane mills (world's largest producer of sugar), sulfitation is the technique widely used for color reduction during the white crystal sugar manufacturing, even though it has many operational limitations, such as environmental problems (for example, atmospheric pollution and acid rain), workplace security, difficulty of process adjustment and sulfur residues in sugar above European, Canadian and North American market legislation. In this way, there are barriers to commercialization of Brazilian crystal white sugar, then Brazil has been exporting basically VHP sugar (very high polarization), which is a raw sugar with low added value (Morilla et al., 2015; Morilla et al., 2016; Hugot, 1986; Rein, 2007). Our group has been evaluating some alternative techniques to replace sulfur

as clarifying agent in the sugarcane juice process, like ozone (Sartori et al., 2017), hydrogen peroxide (Sartori et al., 2017; Mandro et al., 2017) and ionizing radiation (Lima et al., 2016). Electrocoagulation emerges as a possible substitute for sulfitation due to its success in potable water and wastewater treatment (tannery, electroplating, dairy, textile and oil industries) (Chen, 2004; Tak et al., 2015). The process is based on the production of coagulating agents *in situ* by electrical charge; such coagulants can aggregate impurities in the solution and remove them by flotation and/or sedimentation. The kind of coagulants depends on the type of electrode (Garcia-Segura et al., 2017). Aluminum is widely used once it has a low acquisition price, high availability, presents higher effectiveness to remove organic carbon and greater durability than iron (another typical electrode material) (Asha and Kumar, 2016; Souza et al., 2016; Emamjomeh and Sivakumar, 2009). Electrocoagulation with aluminum electrode presents the half-reactions represented by Equations 1, 2 and 3.

$$Al \rightarrow Al^{3+} + 3\ e^{-} \quad (1)$$

$$2\ H_2O + 2e^{-} \rightarrow H_{2\,(g)} + 2\ OH^{-} \quad (2)$$

$$Al^{3+}_{(aq)} + 3\ H_2O \rightarrow Al(OH)_3 + 3\ H^{+}_{(aq)} \quad (3)$$

The half-reaction of Equation 1 happens at the anode when as electrical charge is applied. Meanwhile, Equation 2 occurs at the cathode. The Al^{3+} generated in the solution spontaneously hydrolyses and forms monomeric species ($Al(OH)_3$). Eventually, $Al(OH)_3$ can polymerize in $Al_2(OH)_2^{4+}$, $Al_6(OH)_{15}^{3+}$, $Al_7(OH)_{17}^{4+}$, $Al_8(OH)_{20}^{4+}$, $Al_{13}O_4(OH)_{24}^{7+}$ and $Al_{13}(OH)_{34}^{5+}$, although the main coagulant is $Al(OH)_3$ (Garcia-Segura et al., 2017; Gurses et al., 2002). Electrocoagulation has four most important mechanisms to remove organic material: a) entrapment (trapping impurities with coagulant), b) adsorption (similar to entrapment, however, there are physico-chemical interactions), c) charge neutralization (coagulant blocks the free charges of impurities) and d) complexation (coagulant binds to functional groups of impurities, evolving the molecule) (Nassef, 2014; Garcia-Segura et al., 2017). Those mechanisms normally happen simultaneously and they are efficient to remove impurities

including phenolic compounds as showed by Fajardo et al. (2014) who treated a synthetic mixture of gallic and 4-hydroxybenzoic acids.

Another sugarcane sector lag is the juice color monitoring. In sugarcane mills, the methodology is based on laboratory analysis established by Icumsa (International Commission for Uniform Methods of Sugar Analysis), where absorbance at 420 nm variation is measured (Icumsa, 2011). However, the colorant in sugarcane is very heterogeneous and it is not indicated to be monitored using one wavelength. For example, the reaction of complexation of cathecol and iron that happens in sugarcane juice is not sensitive at 420 nm (Clarke and Godshall, 1988). Icumsa also had an official measurement at 520 nm and there are some works that used this methodology (Chai et al., 2015, Pacheco et al., 2012). Still, there are some recommendations of analysis at 280 nm, once most colorants have their peak at this wavelength (Clarke and Godshall, 1988). Despite their usage, all absorbances mentioned above have problems with influence of pH in the sugarcane juice color and depending on the acidity the solution can be lighter or darker. To date, the use of chromatic variations has been scarcely applied to evaluate sugarcane juice clarification, especially for alternative processes such as electrocoagulation. Considering the demand to replace the current sugarcane juice clarification process (sulfitation) and the need to evolve the color monitoring system during the white crystal sugar manufacturing, this work aimed to evaluate the efficiency of electrocoagulation as an alternative clarification process focusing on the technological parameters normally evaluated in the industry (Icumsa color and turbidity), phenolic compounds content and CIELAB color parameters. Then it was aimed to determine which color analyses are fundamental and critical for monitoring sugarcane changes.

2. Material and Methods

2.1 Sugarcane samples and juice extraction

Sugarcane plants were harvested randomly from Fazenda Areão located in Piracicaba/SP, Brazil (22°43'30" S, 47°38'56" W). Sugarcane juice was obtained from healthy sugarcane plants. They

were disintegrated in forage and pressed (1 min at 250 kgf cm^{-2}) to obtain raw sugarcane juice, which was sifted against sieves at 200 mesh to remove insoluble impurities, sand and fibers. The juice was immediately stored at -18 °C until the electrocoagulation assays.

2.2 Treatment of sugarcane juice by electrocoagulation

Electrocoagulation process was carried out in a 400 mL glass reactor with a jacket. It was coupled to a thermostated water-bath, which kept the reaction temperature constant. A Pump-King (HP 1/30, 1550 rpm, 60 cycles) transferred water from the water-bath to the reactor. The electrodes were two aluminum plates (15 × 3.5 × 2 cm) and were disposed in vertical position with 80% of its area emerged in sugarcane juice. A transformer (Superior Electric, Connecticut, USA) was used to supply energy to electrodes (0-120 V; 15 A) (Figure 1). Four kinetics of color reduction from sugarcane juice were carried out with 300 mL of it. The parameter conditions were set according to preliminary tests and closer to the sugarcane mills'. Temperature, pH, distance between electrodes and soluble solids (Brix) were fixed at 40 ºC (sugarcane juice temperature before entering the treatment during crystal sugar manufacturing), 5.5 (sugarcane juice normal pH), 1 cm (determined in previous tests) and 18º Brix (similar to sugarcane juice). The essays were divided according to the voltage applied: 35, 45, 55 and 65 V (also based on previous tests). The process had a duration of 1 h and 5 mL samples of clarified sugarcane juice were collected every 10 min with a syringe (the process did not need to stop). After that they were centrifuged to remove the supernatant and sludge formed during electrocoagulation. Then the clarified sugarcane juice samples were stored at -18 °C until the analyses of turbidity, Icumsa color, total and simple phenolic compounds and tristimulus colorimetry.

2.3 Technological parameters

Icumsa color and turbidity are parameters normally monitored in the sugarcane mill and both were analyzed as part of technological parameters. To measure Icumsa color, sugarcane juice was diluted to 1.25º Brix. The samples were filtered through PTFE membrane (0.45 µm) and the pH was adjusted to 7.0 using NaOH (0.05 mol L^{-1}). Then the absorbance was measured at 420 nm using

spectrophotometer (UVmini 1240, Shimadzu Co., Japan). The final result was expressed as dimensionless Icumsa color using Equation 4, where: ρ is the samples' density (determined for Equation 5); $Brix_0$ is the initial soluble solids content; $Brix_c$ is the soluble solids content of diluted samples and Abs_{420nm} is absorbance at 420 nm (ICUMSA, 2011).

$$\text{Icumsa color} = [(Abs_{420nm} \times 1000) / (\rho \times (Brix_c/100)] \quad (4)$$

$$\rho = 1 + [((Brix_0 \times (200 + Brix_0)) / 54000) \times (Brix_c/Brix_0)] \quad (5)$$

Turbidity was measured directly in turbidimiter TB-10000 (Tecnopom Co., Brazil) and the equipment expressed the results in nephelometric turbidity units (NTU).

Icumsa color and turbidity of the four treatments (35, 45, 55 and 65 V) were discussed in percentage of reduction. The initial values (at 0 min) were considered 0% of reduction, and then the evolution of both parameters was in function of the beginning.

2.4 Phenolic compounds by UHPLC

2.4.1 Extraction procedure

1 mL of sugarcane juice was acidified to pH 2 with HCl (37% w/v) and the extraction of phenolic compounds was carried out with solid-phase cartridges. It was used cartridges with polypropylene filling (XAD-2 Purified Clean - SPE Tube; Supelco) and methanol as eluent. After that, the material was dried in rotary evaporator. It was added 500 µL of ultrapure water and 500 µL of ethyl acetate. The material was homogenized in ultrasound and centrifuged for 5 min at 17.970 × *g* to collect the supernatants. The procedure was repeated three times. Then, the supernatant was dried in rotary evaporator and the extract was suspended with 200 µL of methanol and filtered at 0.45 µm prior to their injections in the Ultra-High Performance Liquid Chromatography (UHPLC) system.

2.4.2 UHPLC-DAD

Chromatography analyses were performed in duplicate using UHPLC Agilent 1290 system with diode-array detector (DAD), which was set to scan from 200 to 770 nm. The methodology covered

flavonoids and phenolic acids with low molecular weight. The separation was performed using C_{18} Eclipse Plus 120 column (1.8 µm, 50 × 2.1 mm) at 20°C. The mobile phase profile was: formic acid/water at 0.01% proportion (A) and acetonitrile (B). The linear gradient was: 0-5 min: 100% A; 5-20 min: 95% A−5% B; 20−22 min: 50% A−50% B; 22 min: washing and re-equilibration of the column. The injection volume and flow rate were 1 µL and 0.70 mL min^{-1}, respectively. The UV-Vis detector was monitoring 280 nm with the software Open Lab ChemStation. The identification of compounds was executed by external calibration from the areas of the chromatographic peaks obtained by DAD detection at 280 nm (Coyago-Cruz et al., 2018).

2.4.3 Total phenolic compounds by spectrophotometric Folin-Ciocalteu method

Concentration of the total phenolic content was determined by Folin-Ciocalteu reagent according to Singleton and Rossi (1965). Briefly, sugarcane juice collected from the treatment (0.25 mL), Folin-Ciocalteu reagent (1.25 mL) and sodium carbonate 20% (m/v; 3.75 mL) were mixed with distilled water to a total volume of 25 mL. The solution was homogenized and left settling during two hours for reaction and stabilization. Then absorbance was measured at 795 nm in spectrophotometer Hewlett–Packard UV-Vis HP8453 (Palo Alto, CA, USA), using 10 mm path length glass cells and distilled water as reference. The standard curve was made with gallic acid and the results were expressed in mg GAE L^{-1}. As was done previously with turbidity and Icumsa color, the results of total phenolic compounds was discussed in percentage of reduction.

2.5. CIELAB color parameters

The kinetics samples were scanned by CAS-140 spectroradiometer covering visible wavelength of light (380−770 nm). The standard illuminant and observer was D65 and 10°, respectively. A plastic cuvette (45 × 12.5 mm) was used against a reference white background (distilled water). Each sample was read three times consecutively, and then colorimetric measurements were the average. The color parameters of space CIELAB (L* - lightness, a* - green-red axis, b* - blue-yellow

axis, C_{ab}* - chroma and h_{ab} - hue) were obtained directly from the spectroradiometer as described by Gordillo et al. (2015).

Moreover, euclidean distance between two points in three-dimensional space defined by L*, a* and b* were calculated to define the color differences (Equation 6).

$$\Delta E^*_{ab} = [(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2]^{½} \quad (6)$$

2.6. Statistical analysis

Statistica version 8.0 software (Statsoft, 2007) was used for statistical analysis. Univariate analyses (F test, Tukey test and Pearson correlation coefficient) were used to discriminate means of turbidity, Icumsa color, total phenolic compounds and colorimetric parameters in each treatment.

3. Results and discussions

3.1. Turbidity and Icumsa color monitoring

Each result of the four electrocoagulation essays (35, 45, 55 and 65 V essays) was compared. The analysis of variance (ANOVA) was executed to verify the existence of significant difference of Icumsa color and turbidity means between the four essays. Since the F-test was significant, it was carried out a comparison of means test (Tukey test) to identify which means of treatment differ.

Each voltage tested (35, 45, 55 and 65 V essay) reduced turbidity after one hour (Table 1). The turbidity reduction was directly proportional to the tension applied. At the end of 35 V essay, the turbidity reduction was 11.9%. This was the only treatment that did not present significance turbidity reduction ($p < 0.05$) throughout the process. 45, 55 and 65 V essays had significance results and presented 44.6, 83.0 and 99.7% turbidity reduction, respectively. Another tendency was the higher voltage treatments (55 and 65 V) decreased the turbidity faster than lower voltage treatments. 55 and 65 V essays in the first 10 min presented a reduction of 81.3 and 44.8% (both significant at ($p < 0.05$) against 2.4 and 1.1% from 45 and 35 V essays (none of them significant at ($p < 0.05$). Furthermore, 45 V essay presented a significant reduction in the turbidity, only after 40 min of electrocoagulation.

Those results were expected once higher voltage meant more coagulants in solution and higher rates of coagulation. During the electrocoagulation process were formed coagulants in the own site of reaction. Coagulants were electrode material (aluminum) that was electrically dissolved. Cathode generates metallic ion and anode releases hydrogen gas (H_2) which assists the flocculation. Consequently, Chen (2004), Holt et al. (2002), Rein (2007) reported that impurities (non-sugars components, like proteins, ashes, pectins, bagacillo (very small bagasse), waxes and others) were decanted and floated.

Electrocoagulation had a better performance comparing to other sugarcane treatment processes, especially considering the 65 V essay that reduced the turbidity after one hour in 99.7%, which meant a reduction from 1,297 NTU to 3 NTU (data not shown). Rodrigues et al. (2018), using ozone, decreased the turbidity to around 200 NTU (50% reduction). Silva et al. (2015) combined ozone with heat treatment, and reduced from 1,240 NTU to 131 NTU (89% reduction). Hamerski et al. (2011) using carbonation decreased to 40 NTU (88.7% of turbidity). Eggleston et al. (2012) tested the application of lime saccharate and milk of lime in factory trials and reduced to 70 NTU (96% reduction) and 120 NTU (95.6% reduction), respectively. In terms of performance, Nogueira and Venturini-Filho (2007) using ultra and microfiltration showed a superior decrease and obtained a sugarcane juice with 0.33 NTU (99.9% reduction), however there were some problems with fouling that limits the workflow of the technique.

The turbidity reduction obtained by 65 V essay (99.7% reduction) would have great reflection in a sugarcane industry, not only in the quality of the crystal sugar but also in the equipment's durability. Normally, in the crystal sugar manufacturing the sugarcane juice turbidity after the clarification process is around 50 NTU. A sugarcane juice with high turbidity is prone to deposit particles in the evaporator and cause problems with heat transfer. Impurities in sugarcane juice will also disturb other processes such as crystallization and centrifugation. If it persists until the end of the

process, the final product will have problems to reach the necessary clarity (Hugot, 1986; Rein, 2007; Eggleston et al., 2012).

Electrocoagulation treatment also reduced Icumsa color (Table 1). After one hour, 55 and 65 V essays showed reduction of 52.3 and 58.7% in the Icumsa color, respectively. Both values were significative ($p < 0.05$). However, 35 and 45 V essays did not change the color statistically at the end of electrocoagulation process. In addition, 35 V essay showed a fluctuation of Icumsa color over time, including a slight increase after 60 min of 3%.

The reduction of Icumsa color, presented some peculiarities comparing with turbidity. 65 V essay showed a reduction of 73.1% in the Icumsa color after 40 minutes and after that a slight increase in Icumsa color, indicating a stabilization (there was no significance difference at $p < 0.05$ between 40 and 60 min). Meanwhile, 55 V essay showed its greatest Icumsa color reduction after 20 min (55.5%) and a stabilization thereafter.

Saska et al. (2010) working with the conventional techniques used in sugarcane industry: defection (hot liming), sulfitation, single and double carbonation showed an Icumsa color reduction of 35, 47, 44 and 74%. While Rodrigues et al. (2018) using ozone reached 57% Icumsa color reduction. Then, the efficiency of color removing of 85% and 55% reached by 65 and 55 V essays are very promising, especially because this work did not use any flocculant or additives, as is normally used in sugarcane industry to achieve better results. Even though electrolytes can be added to improve results (Tak et al. 2015; Nassef, 2014; Kabdasli et al., 2012; Tena-Zaera et al.; 2007), in this work it was not necessary once sugarcane juice already has a rich electrolytes content (Delgado and Cesar, 1977) and also has phosphoric acid which stimulates the flocculation with the formation of better floc structure (Thai et al., 2016).

The non-application of electrolytes reflects in the costs of the process, then the treatment costs of electrocoagulation system was inferior to chemical coagulation (Moussa et al. 2017). Rodriguez et al. (2007) presented that electrocoagulation was cheaper than application of external coagulants (660€

per year against 1206€ per year) even with the energy costs of electrocoagulation being higher (220€ per year against 106€ per year). Considering the sugarcane industry self-sufficiency in electricity (from the burning of the bagasse in boiler), the electrocoagulation potential is even greater.

The reduction of Icumsa color in 55 V and 65 V essays had very strong positive correlation of Pearson with turbidity (both r were 0.80). 45 V also had positive correlation but less strong (r = 0.66) while 35 V did not presented Pearson correlation. We can conclude that electrocoagulation removes some particles that influence in color and turbidity. Higher voltage had more correlation between Icumsa color and turbidity. Higher voltage meant greater of hydrogen gas in the solution, consequently improving flotation and stirring of the solution. In addition there was an increase of monomeric species of Al^{3+} (more flocs) in the solution, therefore improving the coagulation capacity. More monomeric species stimulated the formation of a network of flocs after aggregation; normally this net has a good shape that improve flocculation (Thai et al., 2016; Nassef, 2014).

3.2. Phenolic compounds: quantification and impact of electrocoagulation

Considering all kinetics, it was quantified three classes of phenolic compounds: benzoic acids (gallic acid, *p*-hydroxybenzoic acid, vanillic acid and syringic acid), hydroxycinnamic acids (*p*-coumaric acid and ferulic acid) and flavones (apigenin, naringenin and quercetin-*O*-glucose) (Table 2; Figure A.1). Other works as Zhao et al. (2008) and Duarte-Almeida et al. (2011) also quantified these classes of phenolic compounds in sugarcane juice.

Electrocoagulation did not affect the concentration of benzoic acid, hydroxycinnamic acid and flavonas constantly. The values for those three fluctuated and did not show a .constant pattern behavior. 65 V essay had stronger reduction of turbidity and Icumsa color over time, however benzoic and hydroxycinnamic acid increased its concentrations after 60 min (from 12.1 mg L^{-1} to 14.3 mg L^{-1} and from 10.5 mg L^{-1} to 11.6 mg L^{-1}). While apigenin reduced from 5.4 mg L^{-1} to 4.1 mg L^{-1}. 55 V essay also had significant reduction of turbidity and Icumsa color and showed an increase of benzoic acid (10.6 mg L^{-1} to 10.8 mg L^{-1}) and a reduction of flavonas (3.1 mg L^{-1} to 2.5 mg L^{-1}), however unlike

what ocurred in the 65 V essay, hydrocinnamic acid decreased after 60 min of 55 V electrocoagulation treatment (from 7.5 mg L^{-1} to 7.2 mg L^{-1}). The only group of phenolic compounds which reduced after the stronger treatments (55 and 65 V) was apigenin, although, the weakest treatment (35 V) presented the biggest reduction of apigenin (from 3.2 mg L^{-1} to 1.4 mg L^{-1}).

Analyzing the phenolic compound singly, the lack of pattern also was presented. For example 65 V essay, initiated with 8.03 mg L^{-1} of *p*-coumaric acid, the concentration increased to 8.63 mg L^{-1} after 30 min, and increased again at the end of the process and the concentration was 8.95 mg L^{-1}. While ferulic acid during the 65 V essay decreased from 2.42 mg L^{-1} to 1.16 mg L^{-1} after 30 min and finished with 2.63 mg L^{-1}.

Moreover, analyzing the concentration of simple phenolic compounds it was not noted a reduction throughout the time as it happened with Icumsa color or turbidity. For example, 65 V essay initiated with 28.0 mg L^{-1} and finished with 29.9 mg L^{-1}, but presented only 24.5 mg L^{-1} after 30 min of treatment. 55 V essay also had a decrease from the beginning to 30 min of treatment (from 22.0 mg L^{-1} to 19.8 mg L^{-1}) and finished with an increase in the concentration of simple phenolic compounds (23.6 mg L^{-1}). The hypothesis for the fluctuation was variation in extraction process and irregular distribution of compounds in sugarcane juice. The unclear reduction/increase also indicated that polymerization did not happen during the process. In the sugarcane juice, simple phenolic compounds only influenced color after bonding with other compounds (for example, amino compounds, proteins, polysaccharides, other phenolic compounds) or with each themselves to form higher molecular weight species. Then the simple phenolic content did not present correlation with Icumsa color.

Then, electrocoagulation was not efficient to reduce simple phenolic compounds and the consequence can be a problem, even not affecting the sugarcane juice color directly. As well starch and organic acids, the persistence of phenolic compounds after the conventional process (clarification with sulfitation) affect the quality of produced sugar (Wojtczak et al., 2014; Wojtczak et al., 2013; Payet et al, 2005; Clarke and Godshall, 1988). Phenolic compounds, for example flavonoids and

hydroxycinnamic acids are precursors of yellow color, while and orange color, respectively, if both persist during the crystal sugar manufacturing as simple phenolic compounds. They cause problems in the maintenance of crystal sugar clarity and, consequently, decrease the shelf life and product value (Clarke and Godshall, 1988; Lee et al. 2018).On the other hand, total phenolic compounds presented a reduction according to the voltage applied. The F-test was significant, and then we compared the means of each essay through Tukey test. As well for turbidity and Icumsa color, total phenolic compounds decreased more in treatments with higher voltage. 65, 55, 45 and 35 V essays after 60 min presented a reduction of 35.2, 27.5, 19.4 and 12.7%, respectively (Table 3). Furthermore, 65 and 55 V essay had a Pearson correlation of 0.94 and 0.85, while 45 V of 81%. It is important to note that in 65 V essay, turbidity was almost zero after a reduction of 99.7%, but Icumsa color had a reduction of 69.3% indicating that colloidal components as phenolic compounds were not removed completely during the process and it was showed as complex phenolic. These results indicated that phenolic compounds influence the sugarcane juice color, especially the complex phenolic compounds (larger chain), once the simple phenolic did not correlate with Icumsa color.

3.3 Chromatic evolution during the electrocoagulation

During electrocoagulation, all treatments had a similar pattern of lightness increase. Essays with higher tension had greater increase in lightness. Those values were proportional with Icumsa color, turbidity and total phenolic compounds, 65 V essay had a higher and faster increase in lightness than other treatments and there were almost no changes in 35 V essay (Figure 2). Therefore L* showed that electrocoagulation accomplished its primary goal of clarifying sugarcane juice. As mentioned above for Icumsa color, higher voltage meant more flocs and more significant increases at lightness (Nassef, 2014).

On the other hand, hue and chroma did not evolve similarly in all treatments. There was the same lightness evolution (increasing throughout the electrocoagulation process) The C^*_{ab} in 35, 45 and 55 V essays showed an increase in the chroma throughout the process and, consequently, Pearson

correlation was really high between L* and C^*_{ab} for those treatments (0.99, 0.98 and 0.94, respectively). However 65 V essay showed a stabilization from 20 min of electrocoagulation process, in this way, this treatment did not show a very high Pearson correlation (0.79) (Figure 3).

For the hue evolution, 35, 45 and 55 V essay almost did not have difference during electrocoagulation with all samples having positive values for a* and b* and remaining in the first quadrant of (a*b*)-plan (Figure 4). However, 65 V had significative change from 50 minutes, once the samples were classified in the first quadrant of (a*b*)-plane in the beginning and, from 50 min, samples were dislocated to second quadrant (a* positive and b* negative) (Figure 4). Changes in h_{ab} indicated differential of pigmentation from 50 min, therefore during 65 V essay some pigments of sugarcane juice could have been coagulated and removed from the clarified sugarcane juice. This conclusion opened a wide horizon because this removal would impact not only in sugarcane juice treatment, but also in the crystal sugar color stabilization, once the composition of pigments influence during storage.

The mean color difference (ΔE^*_{ab}) was also calculated to monitor the electrocoagulation treatments. Values above 3 CIELAB units can be distinguished by human eye (Rodríguez-Pulido et al, 2013). 45, 55 and 65 V essays had a mean color distance between each point of 3.7, 4.2 and 12.1, respectively. All treatments (except for 35 V essay) had visible alteration and were proportional to the applied voltage. During the process 45 V and 55 V essays had similar behavior, however 55 V essay presented faster changes of color. After 10 min the difference was almost 3 times higher for 55 V than 45 V (9.4 against 3.4 CIELAB units), although in the end both had similar values (the difference was only 0.5 CIELAB units). 65 V essay had stronger difference and had three leaps. In the first 10 min the ΔE^*_{ab} was 14.5 CIELAB units, between 20 and 30 min was 14.5 CIELAB unit and between 30 and 40 min the ΔE^*_{ab} was 23.5 CIELAB units. After 30 min in 65 V essay, the hue changes began, while L* was still increasing.

4. Conclusion

The electrocoagulation treatment significantly reduced Icumsa color, turbidity and total phenolic compounds. Higher voltages applied accomplished better results of sugarcane juice clarification. However, all treatments did not influence in simple phenolic content analyzed in this work. Tristimulus analysis presented some pattern that went beyond technological analysis, including that 65 V essay after one hour changed the pigmentation of sugarcane juice and had an early stabilization on chroma. This kind of results could be useful for industry, once they could correlate quality with different color parameters and finally improve the clarification in general with finer settings of technique according to different situations.

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Figures

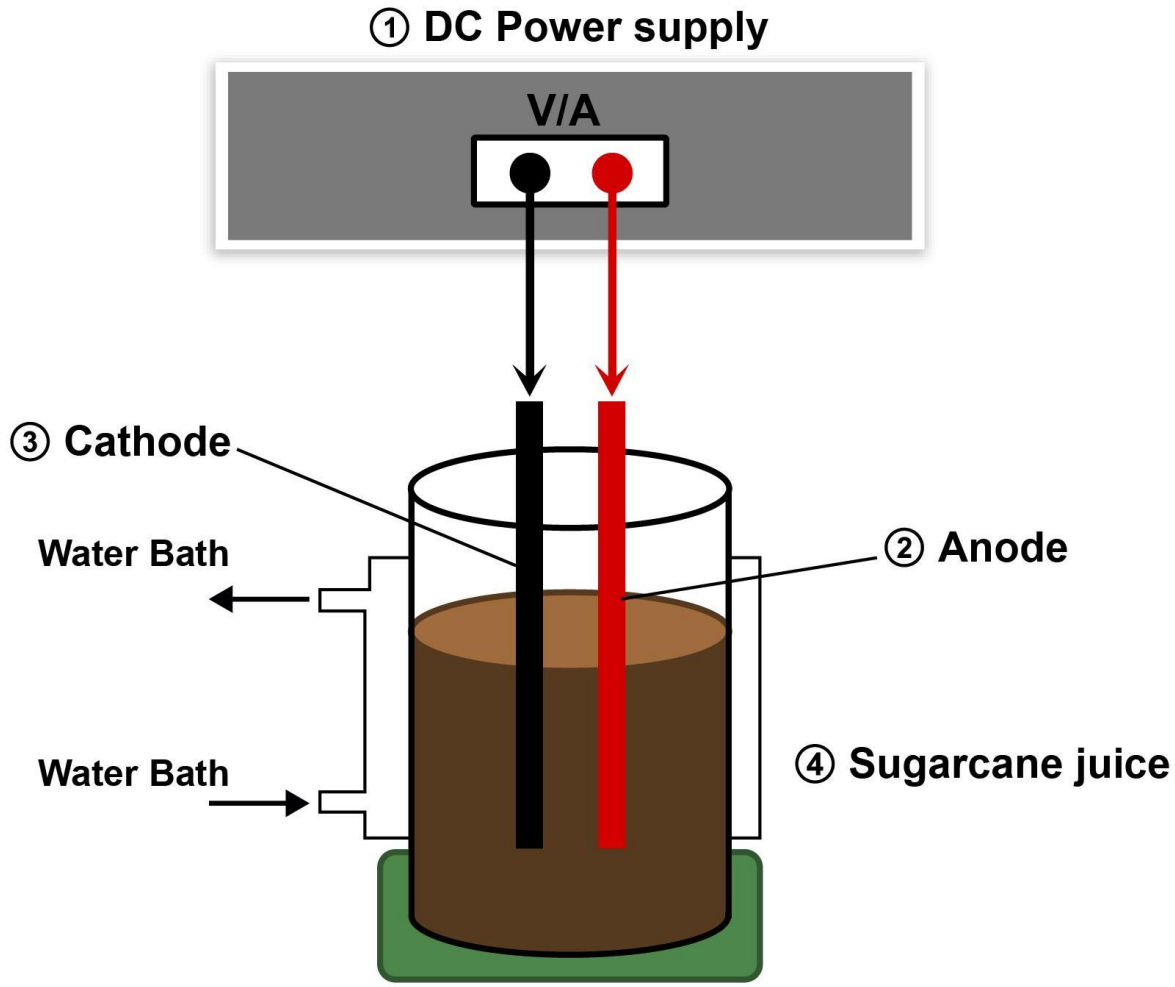


Figure 1. Electrocoagulation glass reactor.

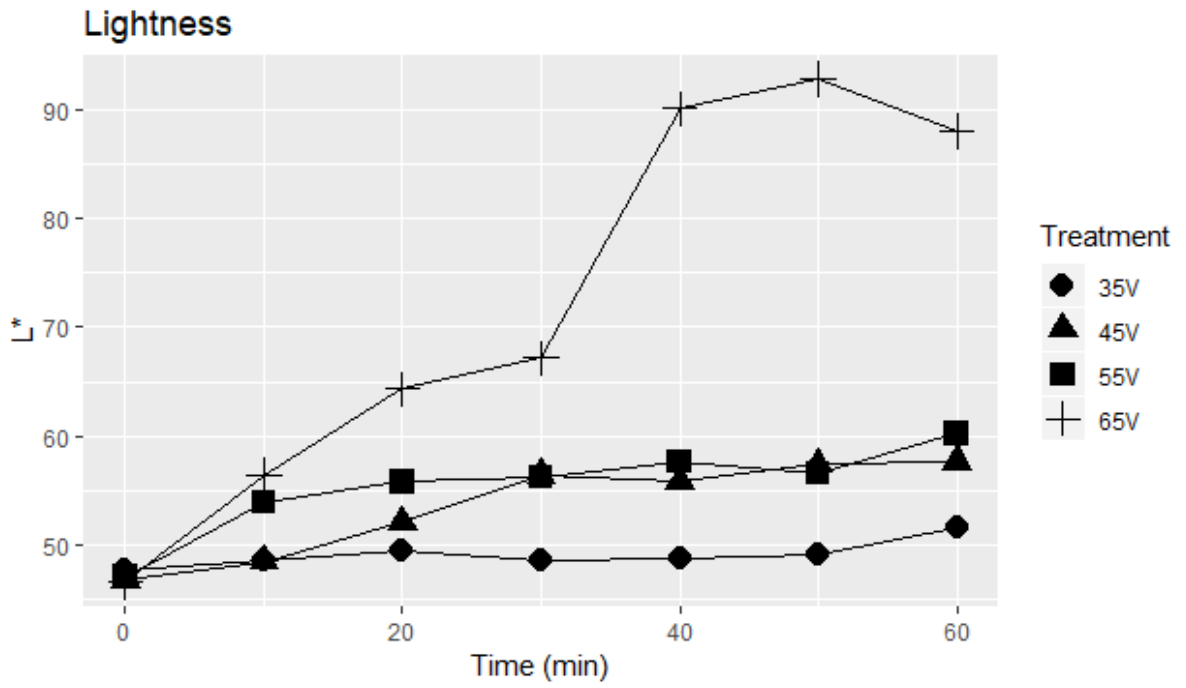


Figure 2. Evolution of lightness parameter of CIELAB (L*) for each treatment.

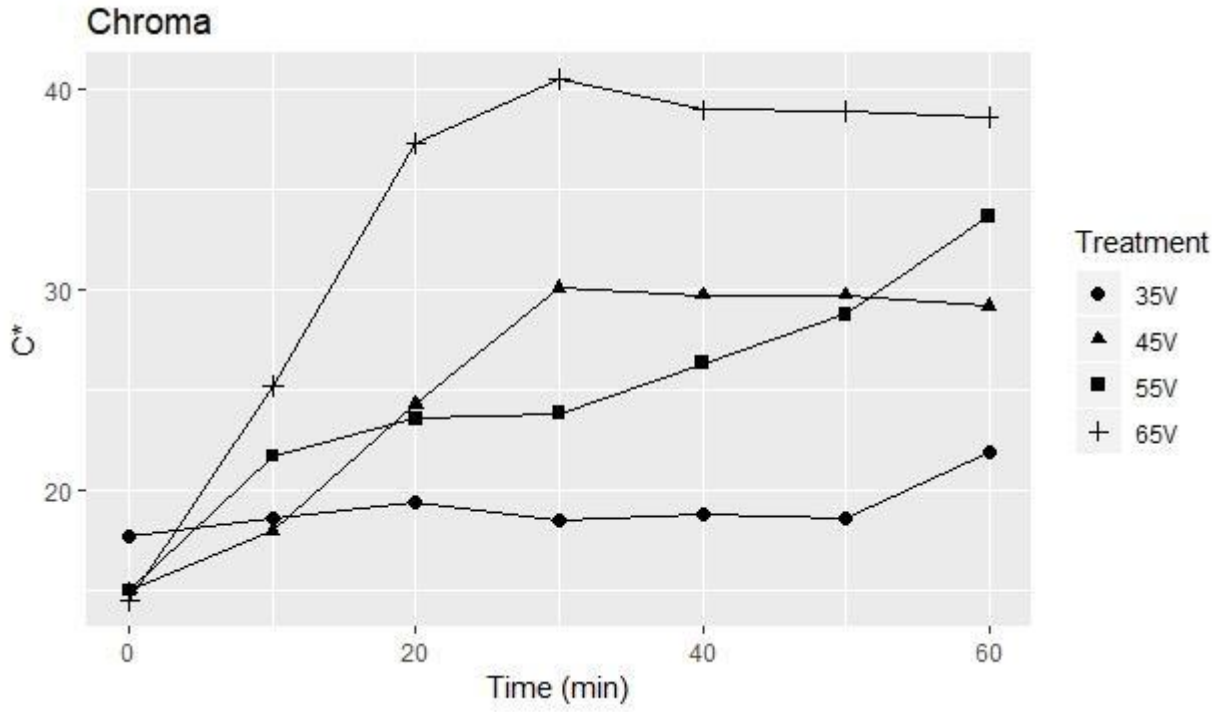
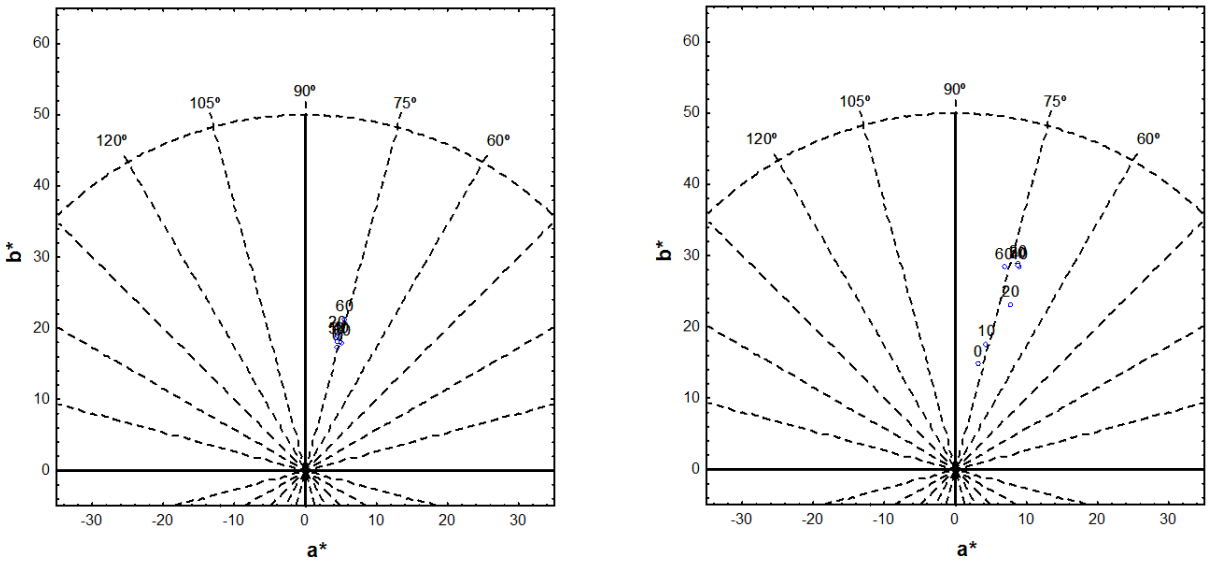
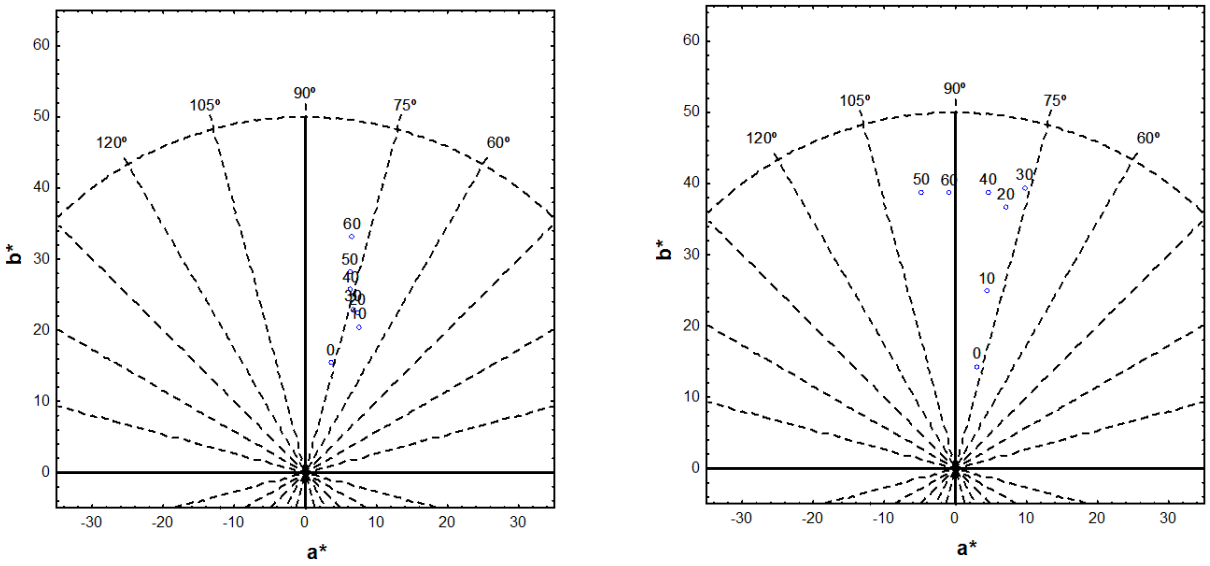


Figure 3. Evolution of chroma parameter of CIELAB (C^*_{ab}) for each treatment.



(35 V) (45 V)



(55 V) (65 V)

Figure 4. CIELAB color space (a*b* - plan) for sugarcane juice treated by electrocoagulation.

Table

Table 1. Icumsa color and turbidity (and its standard deviation) throughout electrocoagulation treatments. The values were in percentage of reduction (%).

Time (min)	Turbidity				Icumsa color			
	35 V	**45 V**	**55 V**	**65 V**	**35 V**	**45 V**	**55 V**	**65 V**
0	0.0% ± 0.438%^{a}	0.0% ± 0.516%^{a}	0.0% ± 0.397%^{a}	0.0% ± 0.749%^{a}	0.0% ± 1.1%^{AB}	0.0% ± 1.1%^{AB}	0.0% ± 1.1%^{AB}	0.0% ± 1.3%^{AB}
10	1.1% ± 0.130%^{a}	2.4% ± 0.348%^{a}	44.8% ± 0.102%^{bcde}	81.3% ± 0.045%^{de}	2.8% ± 11.2%^{ABC}	-3.0% ± 10.3%^{A}	8.8% ± 3.3%^{ABCDE}	10.3% ± 4.6%^{ABCD}
20	3.5% ± 0.085%^{ab}	35.3% ± 0.050%^{abcd}	51.5% ± 0.102%^{bcde}	78.9% ± 0.000%^{de}	0.0% ± 5.9%^{A}	-10.0% ± 3.6%^{A}	52.2% ± 12.2%^{EFG}	39.3% ± 8.3%^{BCDEFG}
30	6.4% ± 0.130%^{abc}	44.9% ± 0.217%^{bcde}	48.7% ± 0.088%^{bcde}	75.7% ± 0.278%^{de}	-8.0% ± 7.2%^{A}	6.9% ± 4.3%^{ABCD}	51.7% ± 1.8%^{EFG}	47.1% ± 10.8%^{DEFG}
40	9.3% ± 0.130%^{abc}	51.5% ± 0.050%^{bcde}	54.9% ± 0.102%^{cde}	99.9% ± 0.001%^{e}	-5.0% ± 4.5%^{A}	16.5% ± 11.5%^{ABCDEF}	39.7% ± 7.2%^{CDEFG}	70.4% ± 1.3%^{G}
50	12.0% ± 0.345%^{abc}	49.3% ± 0.537%^{bcde}	71.4% ± 0.134%^{de}	99.9% ± 0.002%^{e}	-9.0% ± 13.3%^{A}	18.1% ± 3.0%^{ABCDEF}	50.9% ± 12.6%^{EFG}	69.8% ± 5.9%^{G}
60	11.9% ± 0.385%^{abc}	44.6% ± 0.086%^{bcde}	83.0% ± 0.051%^{de}	99.7% ± 0.008%^{e}	-3.0% ± 2.1%^{A}	17.5% ± 40.8%^{ABCDEF}	52.3% ± 14.4%^{EFG}	58.7% ± 1.7%^{FG}

Turbidity values and standard deviation with different lower case are statistically different by Tukey test ($\langle < 0.05$).
Icumsa color values and standard deviation with different capital letter are statistically different by Tukey test ($\langle < 0.05$).

Table 2. Concentration and standard deviation of individual phenolic compounds, benzoic acids, hydroxycinnamic acids, flavonas and simple phenolic expressed in mg L^{-1} identified during the electrocoagulation treatments (35, 45, 55 and 65 V).

	Treatments											
Phen. Comp mg L^{-1}	**35 V**			**45 V**			**55 V**			**65 V**		
	0 min	**30 min**	**60 min**	**0 min**	**30 min**	**60 min**	**0 min**	**30 min**	**60 min**	**0 min**	**30 min**	**60 min**
Gallic acid	1.05± 0.00	1.58± 0.06	1.60± 0.03	1.03± 0.03	0.94± 0.03	1.66± 0.00	0.59± 0.03	0.77± 0.03	0.57± 0.06	0.88± 0.00	0.39± 0.12	1.23± 0.12
p-Hydroxybenzoic acid	1.91± 0.00	1.43± 0.04	1.62± 0.08	1.51± 0.16	1.37± 0.04	1.03± 0.04	1.34± 0.00	1.03± 0.03	0.43± 0.00	1.57± 0.00	1.20± 0.04	0.43± 0.00
Syringic acid	8.06± 0.13	7.08± 0.10	8.35± 0.04	6.13± 0.03	7.24± 0.00	6.36± 0.80	6.07± 0.06	6.93± 1.22	7.26± 0.16	7.83± 0.06	8.35± 0.16	9.89± 0.03
Vanillic acid	3.47± 0.03	2.09± 0.43	2.76± 0.00	1.91± 0.09	1.38± 0.39	1.78± 0.09	1.54± 0.26	0.74± 0.00	5.68± 0.30	1.84± 0.17	0.65± 0.04	2.70± 0.09
∑*Benz acid*	*14.5± 0.16*	*12.2± 0.51*	*14.3± 0.07*	*10.6± 0.29*	*10.9± 0.46*	*10.8± 0.93*	*9.6± 0.33*	*9.5± 1.28*	*13.9± 0.52*	*12.1± 0.23*	*10.6± 0.20*	*14.3± 0.24*
p-Coumaric acid	9.56± 0.06	8.15± 0.04	10.8± 0.11	6.75± 0.18	8.40± 0.05	7.95± 0.37	7.53± 0.18	7.41± 0.56	7.19± 0.11	8.03± 0.00	8.63± 0.20	8.95± 0.06
Ferulic acid	1.87± 0.57	1.26± 0.14	1.06± 0.14	1.87± 0.14	0.00± 0.00	0.00± 0.00	ND	ND	ND	2.42± 0.21	1.16± 0.00	2.63± 0.07
∑*Hydroxyc. acid*	*11.4± 0.62*	*9.4± 0.18*	*11.9± 0.25*	*8.6± 0.33*	*8.4± 0.05*	*7.9± 0.37*	*7.5± 0.18*	*7.4± 0.56*	*7.2± 0.11*	*10.5*± 0.21	*9.8*± 0.20	*11.6*± 0.13
Apigenina	0.85± 0.00	0.00± 0.00	0.00± 0.00	0.00± 0.00	0.32± 0.04	0.00± 0.00	0.73± 0.06	0.41± 0.13	0.00± 0.00	1.64± 0.04	1.03± 0.00	0.00± 0.00
Naringenina	2.35± 0.00	2.31± 0.11	1.36± 1.40	2.37± 0.03	2.45± 0.03	2.29± 0.03	2.37± 0.03	2.5± 0.11	2.45± 0.08	2.99± 0.06	3.14± 0.17	3.01± 0.08
Quercetin-o-glucoside	ND	ND	ND	ND	ND	ND	ND	ND	ND	0.80± 0.02	0.00± 0.00	1.07± 0.00
∑*Flav.*	*3.2± 0.00*	*2.3± 0.00*	*1.4± 1.4*	*2.4± 0.03*	*2.8± 0.07*	*2.3± 0.03*	*3.1± 0.08*	*2.9± 0.24*	*2.5± 0.08*	*5.4*± 0.12	*4.2*± 0.17	*4.1*± 0.08
Simple Phen.	29.1± 0.78	23.9± 0.57	27.5± 0.18	21.6± 0.64	22.1± 0.58	21.1± 0.93	22.0± 0.74	19.8± 2.08	23.6± 0.71	28.0± 0.56	24.5± 0.57	29.9± 0.45

ND - not detected.
∑Benz. acid - sum of concentration of benzoic acids (gallic, p-hydroxybenzoic, vanillic and syringic acids)
∑Hydroxyc. acid - sum of concentration of hydroxycinnamic acids (caffeic, p-Coumaric and ferulic acids)
∑Flav. - sum of concentration of flavonas (apigenina, naringenina and quercetin-o-glucoside)
∑Simple phen - sum of concentration of all individual phenolic compounds identified

Table 3. Concentration and standard deviation of total phenolic compounds in percentage of reduction (%) during electrocoagulation treatment.

Time (min)	Total Phenolic Compounds			
	35 V	**45 V**	**55 V**	**65 V**
0	0.0% ±1.2%^{AB}	0.0% ± 0.6%^{AB}	0.0% ± 0.1%^{AB}	0.0% ± 0.1%^{AB}
10	9.1% ± 1.0%^{ABCD}	-15% ± 2.4%^{A}	11.9% ± 1.8%^{BCDE}	19.2% ± 0.4%^{BCDEF}
20	17.0% ± 0.9%^{BCDEF}	5.5% ± 1.9%^{ABCD}	20.6% ± 1.5%^{BCDEFG}	17.0% ± 2.4%^{BCDEF}
30	12.4% ± 0.6%^{BCDE}	9.7% ± 2.1%^{BCDE}	15.8% ± 1.4%^{BCDEF}	30.1% ± 1.2%^{EFGH}
40	5.5% ± 0.3%^{ABCD}	20.0% ± 2.4%^{BCDEFG}	24.4% ± 2.2%^{CDEFGH}	45.7% ± 1.5%^{H}
50	4.0% ± 0.3%^{ABC}	24.1% ± 0.5%^{CDEFGH}	26.2% ± 0.6%^{CDEFGH}	42.2% ± 2.1%^{GH}
60	12.7% ± 0.6%^{BCDE}	19.4% ± 4.9%^{BCDEF}	27.5% ± 0.5%^{DEFGH}	35.2% ± 1.5%^{FGH}

Total phenolic compounds values and standard deviation with different capital letter are statistically different by Tukey test ($\alpha < 0.05$).