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Incidence of hydrocolloid type on quality parameters in mango leathers
(Mangifera indica L.) Yulima variety
Andrea Milena SÁNCHEZ RIAÑO¹, Helga Patricia BERMEO ANDRADE²,
Claudia Patricia VALENZUELA REAL¹*  

Abstract  
The effect of four hydrocolloids on the physico-chemical, bioactive and rheological properties in sweetened mango leather sheets of the Yulima variety were analyzed. Parametric and non-parametric statistical tests were done to analyze the differences among samples (four of treatment against two of control). The findings show significant positive effects caused by the hydrocolloids made from Gum Arabic (AG), Maltodextrin (MTD) and Citric Slow Pectin (CSP) on leathers’ quality and appearance attributes. Only the Carboxymethylcellulose (CMC) reported adverse effects on the mango leather’s quality; therefore, its use on an industrial scale is not recommended for this line of processed product.  

Keywords: fruit leathering; mango pulp; food additives; polysaccharides; quality attributes.  

Practical Application: Dehydrated and standardized mango leathers made of sweetened mango pulp from Yulima variety, with an alternative selection of hydrocolloids that improve for texture and quality.  

1 Introduction  

Colombia produces a large variety of fruits, mostly seasonal; this means that there are periods of shortages and oversupply during the year. In periods of oversupply, the price of fresh fruit declines by more than 50%, so that in the absence of agroindustrial alternatives to transform the harvest surplus, the profitability of agribusiness declines and product losses should increase until 20-30% of what is produced. This reality is not alien to productive sectors such as mango, which is located in central and caribbean regions of the country (Agronet, 2014). Tolima stands out in the central region as the second producer of this fruit in Colombia, with about 78 thousand tons in year 2014 (Tolima, 2015; Procolombia, 2015), mostly marketed fresh in the absence of agro-industrial alternatives in the region.  

Tolima’s mango producers work with native and improved varieties, including Yulima variety (about 20% of the regional production). This variety is distinguished by a medium-sized and elongated fruit, with yellow to deep red skin, a slight bulge at its apex, low fibre content and bittersweet taste (Asociación Hortífrutícola de Colombia, 2013).  

In regards to its composition, mango (Mangifera indica L.) is a fruit rich in dietary fiber, vitamins, minerals, phenols, carotenoids, tannins, organic acids, sugars and mangiferin, among other components that show nutraceutical and biological activity of interest for human health (Ribeiro & Schieber, 2010; Velderrain-Rodriguez et al., 2016). The products with added value derived from the fruit are diverse: juice, marmalade, nectar, pulp, pickles, snacks and fruit leathers (Jahurul et al., 2015; Oliveira et al., 2012).  

Fruit leathers are the result of processed products made of fruit pulp mixed together with food additives such as carbohydrates (glucose, sorbitol, maltodextrin, gum and pectin) and nutraceuticals compounds (inulin, calcium, vitamins). These compounds define the leather’s instrinsic properties: taste, texture, flexibility, color, viscosity, among others (Akhtar et al., 2014; Singh Gujral & Singh Brar, 2003; Karmas & Karel, 2005; Saha & Bhattacharyya, 2010). Other fruits such as papaya, apple, pomegranate, strawberry, kiwi and grapefruit, have been also reported in the literature as natural raw material to produce fruit leathering (Addai et al., 2016; Pushpa et al., 2006; Tontul & Topuz, 2017; Torres et al., 2015). Results show there are some differences in attributes such as sensory quality and nutritional status, as well as similarities in attributes such as conservation, uses, packaging and storage (Diamante et al., 2014).  

In order to offer a technical alternative to mango producers, in coherence with the regional development policies addressed to strength the agroindustry in Tolima (2016), this research focuses on evaluating the effect of four (4) hydrocolloids on the nutritional attributes and the quality of mango leathers, with the intent of standardizing a product that is new to the region and suitable to the agro-industrial sector.  

2 Materials and methods  

2.1 Plant material and reagents  

Yulima variety mangos of 3/4 ripeness degree were selected for experimentation. The fruits were washed and disinfected with Timsen (1 g/L per 10 min), scalded by water immersion.
Hydrocolloid incidence on mango leather quality

The moisture content (MC) and dry matter (DM) were determined by drying at 70 °C for 24 hours until constant weight according to the method 20013,80 (Association of Official Analytical Chemists, 1980) with some modifications. Prior to drying, the mango leathers (0.5 g) were diluted with 5 mL of deionized water in a double boiler bath (80 ± 0.1 °C) up to water evaporation. The fresh pulp and the pulp with acids were worked with 1 g of sample and the drying was carried out under the same conditions as the fruit leathers.

Color was assessed by means of the CIE L*a*b* system using a colorimeter (Konica Minolta, D65, model CR-410, Tokyo - Japan). Parameters L* (Luminosity and Brightness), a* and b* (chromatic coordinates: Green-red and blue-yellow, respectively), were measured on leather samples of 6 x 6 cm. Parameters C* (color saturation) and angle hue (h: Tonalidad) were calculated according to the equations reported by Ford & Roberts (1998) for system CIE L*a*b*.

2.4 Evaluation of antioxidant capacity

The extracts of mango pulp leathers were obtained using the method described by Hernández & Fernández (2013) with some modifications, 1 g of fruit leather was mixed with ethanol at 96% (in relation 1/5 w/v), using a sequential extraction system until exhausting the sample which involved the following stages: i) Stirring at 300 rpm/1 h / 20 °C (Shaker IKA, IKA Works Inc., KS 4000 IControl, Staufen - Germany); ii) Ultrarruxar for 2 min at 10,000 rpm (IKA, IKA Works Inc., T-25digital, Staufen - Germany); iii) centrifugation at 7,000 rpm/ 20 min (Gemmy, Gemmy Industries, PLC-01, Taipei - Taiwan). The precipitated obtained was discarded and the supernatant was recovered and calibrated to 10 mL with 96% ethanol for further analysis.

The Folin-Ciocalteu method described by Shaghaghi et al. (2008) was followed in order to determine the total phenolic content (TPC), by varying the quantity of the extract or standard sample, implemented with the Folin reagent, from 0.5 mL to 0.25 mL. The obtained and interpolated absorbances in a standard curve of gallic acid allowed to express the results as equivalent milligrams of gallic acid per 100 grams of fruit leather (mg GAE/100 g FL).

The colorimetric method of total antioxidant capacity -ABTS radical•+ described by Kuskoski et al. (2005), was applied with some modifications: (i) 3.43 mL of ABTS•+ radical (absorbance adjusted to 0.700 ± 0.100 with ethanol at 96%) mixed in a quartz cell with 70 µL of ethanolic extract at different concentrations and ii) Control of absorbance at 754 nm at 6 min of reaction. Trolox calibration curve was set up under the same conditions and the results were expressed as Trolox equivalent antioxidant capacity (TEAC - mM/100 g LF).

For the determination of the stabilizing capacity of the DPPH• Radical the methodology of Kuskoski et al. (2005) was followed with some adjustments. 88 µL of each extract at different concentrations was mixed with 3.412 mL of radical DPPH• (100 µM in methanol), in test tube isolated from light and oxygen. At 30 min of reaction was read the absorbance in a spectrophotometer at 517 nm. A Trolox curve, a control without sample and a solvents blank were estimated under

2.3 Physicochemical characterization

The parameters were measured as follow: pH with a potentiometer (SI Analytics, Xylem Inc, HL 100, Weilheim - Germany); The TSSC determination with a digital refractometer (Atago, CO. Ltda., PAL-S, Tokyo - Japan); Total Titratable Acidity (TTA), by means of titration with NaD H 0.1 N up to pH 8.2, expressed as citric acid percentage; Reducing Sugar Content (RSC) by spectrophotometry at 540 nm (Genesys, 10S UV-Vis, Thermo Scientific, Massachusetts - USA), in accordance with Miller (1959) method, the results were expressed in milligrams of glucose equivalents per gram of the fruit leather or fresh pulp (mg GE/ g LF or PF).

2.2 Pulp leather preparation

Four texture improving agents were used to produce mango leather sheets: a) Arabic Gum (AG); b) Maltodextrin (MTD); c) Citric Slow Pectin (CSP); and (d) Carboxymethylcellulose (CMC), selected due to their easy industrial access and beneficial results from previous studies. The hydrocolloid concentration was equivalent to 0.25% (p/p) with respect to the total soluble solids content (TSSC) of the formulation, considering the high pectin content present in the fruit (1.23%). The standardized process focused on adjusting the concentration of Total Soluble Solids Content (TSSC) of the pulp up to 25 °Brix with addition of sucrose. Then, the sweetened pulp were heated to 60 °C to incorporate hydrocolloids and preservatives (potassium sorbate 50 ppm). Heat was maintained for 15 minutes to allow the dispersion and dissolution of components as well as to reduce the microbial load and enzymes inactivation. Once the heat treatment was concluded, the mixture was deposited in acrylic plates coated with fibre-reinforced plastic (Cristalplast) (9 ± 1 mm thick) and dehydrated by forced air circulation (Kryove, DiEs, 20-60 °C, Medellin - Colombia) to 50 °C until exhausting the sample which involved the following stages: i) Stirring at 300 rpm/1 h / 20 °C (Shaker IKA, IKA Works Inc., KS 4000 IControl, Staufen - Germany); ii) Ultrarruxar for 2 min at 10,000 rpm (IKA, IKA Works Inc., T-25digital, Staufen - Germany); iii) centrifugation at 7,000 rpm/ 20 min (Gemmy, Gemmy Industries, PLC-01, Taipei - Taiwan). The precipitated obtained was discarded and the supernatant was recovered and calibrated to 10 mL with 96% ethanol for further analysis.

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2.2’azinobis (3- etilbenzotiazolin 6-ácido sulfónico), 1,1-diphenyl-2- picrylhydrazyl, 3,5-Dinitrosaliclyc acid, y (±)-6-Hydroxy-2,5,7,8-tetramethylchromane-2-carboxylic acid from Sigma Aldrich Inc. (St. Louis, USA). Potassium Sorbate (PS), Citric Acid (CA), Ascorbic Acid (AA), Maltodextrin (MTD), Citric Slow Pectin (CSP), Arabic Gum (AG) and Carboxymethylcellulose (CMC) food grade were acquired in Tecnas S.A.S and Cimpa S.A.S. (Bogota, Colombia).

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2.5 Rheological behavior

Viscosity

The different pulp mixes prior to the drying process were evaluated, with the use of a viscometer (Brookfield, Engineering Laboratory, Model DV-II, Massachusetts - USA) and taking as a reference Saha & Bhattacharya (2010) methodology. Samples of 50 g at room temperature of 25 ± 2 °C were placed in a cylinder of that capacity and assessed with use of a spindle LV4. Measurements were taken at 3, 10, 30, 50, 70 and 90 rpm, leaving a stabilization time of 5 min to 0.5 rpm, with a subsequent adjustment of the speed at corresponding measurement value. The viscosity reading (Pa.s) for each measurement speed was recorded at 2 min of the spindle rotation.

Texture

For tension texture test, the methodology described by Singh Gujral & Singh Brar (2003) was followed including some modifications. Rectangular strips of 90 × 18 mm and thickness average of 2 ± 0.2 mm were cut from the center of the pulp leathers, considering 10 replicas per sample. It was used a Universal Testing Machine (Lloyd, Model LS1/TA, from Ametek Inc., Pennsylvania - USA) equipped with a 1 kN cell load. The measurements were carried out at room temperature (24 ± 1 °C), leaving an initial distance between clamps of 50 mm and the crosshead speed was set at 50 mm/min. Only strips broken in half were considered. The rupture strength (N), tensile strength (MPa), extensibility (mm), elongation at break (% E) and Modulus of elasticity (MPa) were calculated using NEXYGENPlus* Software Materials Testing Software 3.0 (Ametek Inc., Pennsylvania - USA).

2.6 Statistical analysis

A descriptive statistical analysis was carried out upon the obtained data for each test that was run. The test were triplicated (n = 3), except for the texture test. Once the data’s assumption of normality was checked, it was applied the analysis of variance (ANOVA) parametric statistical test with multiple comparison Fisher tests ranges (Least Significance Difference - LSD) to estimate significant differences between samples, and Pearson correlation tests were implemented to evaluate the relationship between the antioxidant capacity parameters. For the data that did not follow a normal distribution, the Kruskal-Wallis test was applied together with the box and whisker plot in order to estimate significant differences between medians. In all cases, it was implemented a significance level α = 0.05, by means of using the statistical software Statgraphics*.

3 Results and discussion

3.1 Physico-chemical properties

The results of the physico-chemical characterization recorded in Table 1, allow to see that the pulp leathers differ significantly (p < 0.05) in all the parameters evaluated with respect to the fresh pulp and in most of the attributes of control sample C2. There is evidence of an average increase in the TSSC of the 43% and 57% with respect to the sweetened pulp and fresh fruit respectively, with an increase of RSC possibly by sucrose inversion, given the acid concentrations and heat treatments. The chemical change that was reflected in sensory analysis by means of the high sweetness of pulp leathering, induces to take into account an adequate balance between sugar-acidity, and ripeness degree.

In general, the leathers presented a higher concentration of TSSC, RSC, DM and TTA, with a decrease in MC and an increase in pH. Behavior according to expectations, since the dehydration processes in food matrices increase the ashes and dry matter, in response to removal of the free water present in the fresh material (Ozgur et al., 2011). However, the moisture difference between samples indicates that the type of hydrocolloid affects the % of water removal and retention that affect the pulp leather’s flexibility.

Therefore, the MC that ranged between 14.08 and 22.40%, congruent with reported previous mango studies (Azeredo et al., 2006; Mir & Nath, 1995), as well as a pH of 3.50 present on C1 and hydrocolloids treatments; allows to infer that these products are congruent with reported previous mango studies (Azeredo et al., 2006; Mir & Nath, 1995), as well as a pH of 3.50 present on C1 and hydrocolloids treatments; allows to infer that these products are biologically safe (Knechtges; 2012; Azeredo et al., 2006; Hernández & Fernández, 2013). The order in which the leathers flexibility

Table 1. Physicochemical characterization of ‘Yulima’ mango pulp and leathers.

| Sample   | pH      | TSSC   | TTA      | RSC      | MC        | DM      |
|----------|---------|--------|----------|----------|-----------|---------|
| MTD      | 3.53 ± 0.01a | 69.50 ± 1.30b | 2.52 ± 0.02c | 107.74 ± 1.33d | 20.86 ± 0.28e | 79.14 ± 0.28f |
| CSP      | 3.54 ± 0.00d | 68.50 ± 0.90c | 2.50 ± 0.01b | 111.29 ± 3.04d | 17.65 ± 0.59e | 82.35 ± 0.59f |
| CMC      | 3.55 ± 0.01e | 66.50 ± 0.50f | 2.25 ± 0.01b | 118.34 ± 0.89g | 16.73 ± 0.39h | 83.27 ± 0.39i |
| AG       | 3.54 ± 0.00c | 67.80 ± 0.80c | 2.42 ± 0.02b | 107.65 ± 4.98c | 22.40 ± 0.03d | 77.60 ± 0.03e |
| C1       | 3.53 ± 0.00g | 66.70 ± 1.30d | 2.44 ± 0.03b | 117.36 ± 1.97f | 20.10 ± 0.52g | 79.90 ± 0.52h |
| C2       | 3.48 ± 0.01a | 67.70 ± 0.30a | 5.35 ± 0.34d | 224.61 ± 1.97f | 14.08 ± 0.73c | 85.92 ± 0.73d |
| Fresh pulp | 3.60 ± 0.02h | 10.30 ± 0.10e | 1.08 ± 0.00 | 17.00 ± 0.12f | 84.60 ± 0.06 | 15.40 ± 0.08 |
| Pulp with acids | 3.48 ± 0.01a | 10.70 ± 0.10 | 1.20 ± 0.00 | 19.23 ± 0.39 | 84.30 ± 0.06 | 15.70 ± 0.06 |

pH in solution at 10% for all samples. Total Soluble Solids Content (TSSC: °Brix); Total titulable acidity (TTA: % Citric Acid); Reducing Sugar Content (RSC: mg Glucose Equivalent/g Fruit Leather or Fruit Pulp); Moisture Content (MC: %); Dry matter (DM: %); Maltodextrin (MTD); Citric Slow Pectin (CSP); Carboxymethylcellulose (CMC); Arabic Gum (AG); C1 and C2 are control samples. Values are expressed as ± ds (n = 3). Values with the same letter by parameter do not show significant differences (p > 0.05; Model Least Significance Difference; α = 0.05 %).
is positively affected is AG > MTD > C1 > CSP > CMC > C2, results that highlight an AG and MTD as the best hydrocolloids used to preserve leathers’ physico-chemical quality.

3.2 Color analysis

As a response to the presence of carotenoids, mango pulp at consumption ripeness is characterized by displaying orange and yellow hues produced by α and β-carotene respectively, being the latter being predominant in the fruit (Khoo et al., 2011). This characteristic, except in C2 (Brown color. L*: 10.08; C*: 17.39; h: 54.55), remained in all the analyzed leathers, with L*, C* and h values between 40.62-45.41, 22.16-28.18 and 57.18-60.31, respectively (see Table 2). Luminosity variation (L*) correlates with the degree and increase of food browning (Huang & Hsieh, 2005), as associated with the Maillard reaction (Tamanna & Mahmood, 2015). In this sense, significant color variations were shown (p < 0.05) in all parameters, noting that luminosity / brightness (L*: 0-100), color saturation (C*: 0-150) and tonality (h: 0-360°) resulted favorable, mainly on CSP, AG and MTD.

The color results reveal, firstly, the protective and potential function of the incorporated antioxidants and sugars, on the leathers’ colors (Ali et al., 2015; Clemens et al., 2016); secondly, the strongly influence of hydrocolloid and its interaction with other components contained within the food matrix. Hydrocolloids such as MTD, CSP and CMC have shown delay on non-enzymatic browning reactions and sugar crystalization (Eduardo et al., 2016; Karmas & Karel, 2005), this behavior is attributable to the amorphous matrix as well as the reactants persistent dilution in a highly viscous environment (Buera et al., 2005; Singh Gujral & Singh Brar, 2003).

3.3 Total Phenolic Content (TPC)

The leathers of MTD, CSP, AG and C1 with TPC between 520.4-546.5 (mg GAE/100 g FL) did not show significant statistical differences among them (p > 0.05) (see Figure 1), nevertheless AG showed greater TPC. Beside CMC and C2 displayed significantly different from the rest formulations, revealing minor and major TPC, respectively (455.7 and 1,453.5 mg GAE/100 g FL). It can be noted that C2 was triplicated in this parameter, as opposed to CMC. Ot can be noted that pasteurization is a classic process to regulate the microbial load, deactivate enzymes, increase shelf life and foods quality (Silva et al., 2014).

Rababah et al. (2005) verified that the drying and ascorbic acid addition process did not affect the content of TPC in fruit purée. On the opposite, the phenolic concentration increases considerably in dehydrated product (Slatnar et al., 2011). However, the thermal treatment of pulp like in jam process degraded the phenolic compounds reducing TPC (Rababah et al., 2011), which could explain the results on mango leathers, where all the samples, except those of C2, were pasteurized before the drying process. This highlighted the positive effect of AG on leathers TPC, as opposed to CMC. It can be noted that pasteurization is a classic and effective process to regulate the microbial load, deactivate enzymes, increase shelf life and foods quality (Silva et al., 2014).

3.4 Antioxidant capacity analysis

There is a high correlation between TPC and the capacity of antioxidant leathers, measured as capacity to capture ABTS•+ (0.759) and DPPH• (0.704). Even though these values were quite similar, C2 and CMC showed greater TEAC (p < 0.05) than other leathers (ABTS: 17.0 and 15.2; DPPH: 18.6 and 16.3 mM/100 g FL, respectively) (see Figure 2). This behavior in the leather antioxidant

Table 2. Effect of hydrocolloid type on the color of Yulima mango leathers.

| Sample | a*  | b*  | L*  | C*  | h   |
|--------|-----|-----|-----|-----|-----|
| MTD    | 12.94 ± 0.04± | 21.94 ± 0.02± | 44.47 ± 0.01± | 25.47 ± 0.00± | 59.48 ± 0.09± |
| CSP    | 13.96 ± 0.01± | 24.48 ± 0.01± | 45.41 ± 0.01± | 28.18 ± 0.01± | 60.31 ± 0.02± |
| CMC    | 12.10 ± 0.03± | 18.57 ± 0.01± | 40.62 ± 0.01± | 22.16 ± 0.02± | 56.91 ± 0.05± |
| AG     | 12.83 ± 0.01± | 22.26 ± 0.00± | 43.82 ± 0.01± | 25.69 ± 0.01± | 60.05 ± 0.02± |
| C1     | 12.87 ± 0.01± | 19.96 ± 0.02± | 42.17 ± 0.01± | 23.75 ± 0.01± | 57.18 ± 0.02± |
| C2     | 10.08 ± 0.02± | 14.16 ± 0.01± | 38.53 ± 0.01± | 17.39 ± 0.01± | 54.55 ± 0.05± |

Maltodextrin (MTD); Pectin (CSP); Carboxymethylcellulose (CMC); Arabic Gum (AG); C1 and C2 are control samples. Values are expressed as X ± ds (n = 3). Values with the same letter by parameter do not show significant differences (p > 0.05; Model Least Significance Difference; α = 0.05 %).

Figure 1. Total Phenolic Content of Yulima mango leathers, in presence and absence of hydrocolloids. Maltodextrin (MTD); Citric Slow Pectin (CSP); Carboxymethylcellulose (CMC); Arabic Gum (AG); C1 and C2 are control samples. Values are expressed as X ± ds n = 3.
capacity contrasts with the evidence of TPC, despite the high correlations between the parameters.

The ABTS test, due to its chemical bipolarity, tells about plant extracts’ total antioxidant capacity (Dong et al., 2015), whereas the stabilizing ability of the DPPH radical is often correlated with the ascorbic acid concentration, which in presence of citric acid, improves the stabilizing action of the radical (Scalzo, 2008). It is presumed that the partial thermal degradation of phenolic compounds in the mango pasteurized leathers, and the addition of citric and ascorbic acid in the formulation, was able to generate a synergistic effect-compensatory between acids and antioxidant metabolites from the pulp (Machado et al., 2007; Noguer et al., 2014). As evident in C2 that lacked the addition of acids, and its antioxidant capacity values did differ from the other samples.

### 3.5 Rheological behavior

#### Viscosity analysis

The viscosity decreased with the increase of the shear rate in all formulations, showing a shear thinning behavior of non-Newtonian fluids, with significant influence of the type of hydrocolloid in viscosity (see Table 3). As expected CMC and MTD conferred a higher viscosity to the food matrix, in contrast to AG (Castro et al., 2016; Saha & Bhattacharya, 2010). C2 presented the lowest viscosity to contain only hydrocolloids and sugars presented in fresh fruit (Clemens et al., 2016; Saha & Bhattacharya, 2010), and CSP generated a high viscosity, attributable to its association with sugars as glucose and sucrose in an acid medium, which decrease the dielectric constant of the solvent, favoring the action of dehydration and the formation of hydrogen bonds (Lopes & Rao, 2006).

#### Texture analysis

The fruit leathers, which are characterized by a chewy texture, revealed significant differences (p < 0.05) in the evaluated mechanical parameters, with high influence of hydrocolloid type (see Table 4). The tensile strength or resistance to breakage increased as CSP ≤ AG ≤ C1 ≤ MTD < CMC < C2. Such a trend is closely correlated with the modulus of elasticity, which measures...
the material rigidity (Bourne, 2002), and for mango leathers was kept between 1.36 and 3.05 MPa. AG and CMC conferred greater and lesser extensibility to the mango leathers in correlation with their %E (14.45 mm, 28.90%; 8.82 mm, 17.63%).

The beneficial effect of sugars on the texture and its synergism with CSP, MTD, AG, C1 can be highlighted. According to Singh Gujral & Singh Brar (2003), mango leathering with concentrations of CMC, AG and Pectin (1-3%), are less resistant to rupture (3.7-12.5 N) and more elastic (module of elasticity: 0.23-0.68 MPa). Divergent results with this study may be due to mango variety, ripeness degree, formulation, preparation process of the leathers, strength test and moisture content of the samples.

4 Conclusions

This study revealed the direct influence of hydrocolloid type on the properties and quality of Yulima mango leathers, and its synergy with the components present in the food matrix. Considering the desired attributes in this line of processed product, the physico-chemical parameters evaluated highlighted AG and MTD leathers. The Luminosity/Brightness (L*), saturation (C*) and hue angle, highlighted the protective function and anti-browning of CSP, AG and MTD on the orange-yellow color characteristic of the pulp. Pasteurization used prior to the drying operation significantly reduces the TPC, however, this loss is compensated by the addition and joint action of ascorbic acid and citric acid mainly in leathers' laminates of AG, MTD and CSP. Although the viscosity and texture of the samples differ significantly depending on the type of hydrocolloid, only CMC appears to negatively influence the quality of the final product.

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