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Quality changes of pasteurised orange juice during storage: A kinetic study of specific parameters and their relation to colour instability

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ABSTRACT

In view of understanding colour instability of pasteurised orange juice during storage, to the best of our knowledge, this study reports for the first time in a systematic and quantitative way on a range of changes in specific quality parameters as a function of time and as well as temperature (20–42 °C). A zero-order (°Brix, fructose, glucose), a first-order (vitamin C), a second-order (sucrose) and a fractional conversion model (oxygen) were selected to model the evolution of the parameters between parentheses. Activation energies ranged from 22 to 136 kJ mol^{-1}, HMF formation being the most temperature sensitive. High correlations were found between sugars, ascorbic acid, their degradation products (furfural and HMF) and total colour difference (ΔE^*). Based on PLS regression, the importance of the quality parameters for colour degradation was ranked relatively among each other: the acid-catalysed degradation of sugars and ascorbic acid degradation reactions appeared to be important for browning development in pasteurised orange juice during ambient storage.

1. Introduction

Orange juice is known as the most popular fruit juice worldwide. Although its consumption is decreasing in developed countries, such as USA and Western Europe, in emerging countries, the consumption of juice, along with nectar and still drink, shows an increasing trend (Neves, Trombin, Lopes, Kalaki, & Milan, 2011). Orange juice is highly valued by consumers for its nutritional content, appealing colour and refreshing sweet and sour taste. Among different processing technologies, heat pasteurisation is the most common technique to extend the shelf-life of orange juice. The inactivation of spoilage microorganisms and thermally resistant endogenous enzymes (e.g., pectin methylesterase (PME)) can be achieved under temperature conditions at 90–98 °C for 10–60 s (Vervoort et al., 2011; Yeom, Zhang, & Chism, 2002). From microbial safety point of view, given the fact that orange juice is categorised as high acid food (pH < 4.6), intensively pasteurised orange juice is stable at room temperature (Silva & Gibbs, 2004). However, during storage, as the quality of most food products decreases, changes in sensorial and nutritional qualities of orange juice are the limiting factors determining the 'best before date' listed on the product.

An important quality loss of orange juice during shelf-life is the change in colour. It is the first visible sign which can negatively influence consumers' acceptance, thereby decreasing its commercial value (Manso, Oliveira, Oliveira, & Frías, 2001). It is known that colour change is an indicator for chemical and biochemical reactions (van Boekel, 2008). Depending on the extent of the reaction, changes in colour may occur as a result of the formation of brown pigments as well as the fading of carotenoids, the naturally

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occurring pigments of orange juice. Recently, a kinetic study reported on the carotenoid stability in orange juice and pointed out to their potentially limited contribution to colour change (Wibowo et al., 2015). In intensively pasteurised products, in which quality-degrading enzymes are inactivated, non-enzymatic browning reactions rather than enzymatic browning should be focussed on. Authors have been suggesting different mechanisms for non-enzymatic browning in pasteurised citrus juice: (i) degradation of ascorbic acid (Kaanane, Kane, & Labuza, 1988; Roig, Bello, Rivera, & Kennedy, 1999; Solomon, Svanberg, & Sahlstrom, 1995), (ii) acid-catalysed degradation of sugars (Lee & Nagy, 1988b; Roig et al., 1999) and (iii) Maillard reactions between reducing sugars and amino acids (Bacigalupi et al., 2013; Lee & Nagy, 1988a). Nevertheless, due to the complexity of colour degradation, there is probably more than one mechanism involved. Moreover, interaction between different pathways or compounds and the sequence of occurring reactions could be important in explaining this browning phenomenon. Although some authors have reported on the kinetics of colour change and some of the markers for browning (ascorbic acid, 5-hydroxymethylfurfural or HMF, furfural, etc.) (Bacigalupi et al., 2013; Burdurlu, Koca, & Karadeniz, 2006; Kaanane et al., 1988; Van Bree et al., 2012), there is still a need for a systematic, comprehensive and quantitative study which investigates the change in a range specific quality parameters for colour change under a broad window of shelf-life time and temperature conditions. In addition, the potential of integrating information obtained from multiple responses and the potential to select significant response changes by the use of multivariate data analysis should not be forgotten (Grauwet, Vervoort, Colle, Van Loey, & Hendrickx, 2014).

The objective of the current research paper can be summarised as the study of chemical parameters linked to non-enzymatic browning of heat pasteurised orange juice as a function of storage. Changes in acids (Section 3.1.1), sugars (Section 3.1.2), oxygen (Section 3.1.3), vitamin C (Section 3.1.4), furfural and HMF (Section 3.1.5) were quantitatively studied as a function of storage time and temperature (20 °C, 28 °C, 35 °C and 42 °C). Regarding to this, two steps were taken. First, to quantitatively describe changes of different parameters, a kinetic modelling approach was used for monitoring quality changes during storage as a function of controlled time and temperature conditions (Section 3.1). Secondly, to interpret the mutual importance of the selected quality parameters and their correlation to colour change, multivariate data analysis techniques were applied (Section 3.2).

2. Materials and methods

2.1. Sample preparation

Frozen orange juice concentrate (*Citrus sinensis* (L.) Osbeck) (65 °Brix), purchased from a commercial juice producer in Brazil, was mixed with water at a ratio 1:5 (w/w) yielding 11.2 °Brix$_c$. The single strength juice was pasteurised in a tubular heat exchanger at 92 °C for 30 s. Under this processing condition, no residual pectin methylesterase (PME) activity was determined. Subsequently, juice was hot filled at 85 °C into 500 mL polyethylene terephthalate (PET) bottles and sealed by cap twist inversion. Finally, the bottles were cooled to ambient temperature by submerging them into a tank with circulating chlorinated water. Bottles were stored protected from light at 20 °C and 28 °C for 32 weeks, at 35 °C for 12 weeks and at 42 °C for 8 weeks, in T-controlled incubators (IPP500, Memmert, Schwabach, Germany). At a specific sampling time, bottles were randomly sampled from the incubator, transferred to smaller tubes (±30 mL) and stored at −80 °C. Each tube was thawed in a circulating water bath at 25 °C and homogenised prior to analysis.

2.2. Determination of the acidity

2.2.1. pH and titratable acidity

The orange juice pH was measured at 22 ± 1 °C using a pH meter (Meterlab PHM210, Radiometer Analytical, Villeurbanne, France), which was calibrated with calibration buffers of pH 4.0 and 7.0 (IUPAC, Radiometer Analytical, Villeurbanne, France). All measurements were done in triplicate. Titratable acidity, expressed in percent citric acid, was determined according to AOAC method 962.12 (AOAC, 1998). Analysis was done in triplicate by titrating 10 g of orange juice with 0.1 N NaOH to pH 8.2. The % citric acid was calculated as expressed by Eq. (1).

$$\%\ \text{citric acid(w/w)} = \frac{\text{volume NaOH(ml)} \times 0.64}{\text{juice weight(g)}} \times 100 \quad (1)$$

2.2.2. Organic acid profile

Organic acids were extracted according to the method of Vervoort et al. (2011). 10 mL of orange juice was transferred into a Nalgene centrifuge tube (50 mL). 500 μL of each Carrez I (15% w/v $K_4[Fe(CN)_6]$) and Carrez II (30% w/v $ZnSO_4$) was added and the mixture was homogenised using a vortex mixer. After resting for 30 min at room temperature, samples were centrifuged at 24000*g* for 15 min at 4 °C (J2-HS centrifuge, Beckman, Brea, CA, US). The obtained supernatant was filtered through a 0.45 μm syringe filter (Chromafil A-45/25, Macherey–Nagel, Düren, Germany). 2 μL of the organic acids extract was injected into the reversed phase (RP) HPLC system (Agilent 1200 series, Diegem, Belgium) and separated on a Prevail Organic Acid column (250 mm × 4.6 mm, 5 μm particle size, Alltech Grace, Deerfield, IL) protected with a Prevail C_{18} guard cartridge (7.5 mm × 4.6 mm, 5 μm particle size, Alltech Grace, Deerfield, IL). Isocratic elution (25 mM potassium dihydrogen phosphate buffer pH 2.5) with a flow rate of 1 mL min^{-1} at 25 °C was used. The UV-DAD detector was set at 210 nm. All samples were analysed in triplicates. Identification was done by comparing the retention time and UV spectra with standard solutions of a wide range of organic acids in milli-Q water. Quantification was performed using calibration curves based on the peak area and known concentrations of injected standard solutions, with regression equation for citric acid ($y = 64.32x - 1.53$, $R^2 = 0.99$) and for malic acid ($y = 48.55x + 0.53$, $R^2 = 0.99$).

2.3. Sugar content determination

2.3.1. Total sugar content

The total sugar content, expressed in °Brix was measured in triplicate using a digital refractometer (RX-7000α, Atago, Tokyo, Japan) at 20 °C. The soluble solids (SS) content was determined and expressed in °Brix$_c$, after correction with acid correction factors (Kimball, 1991).

2.3.2. Sugar profile

Sugar extraction and chromatographic analysis were based on the procedure of Vervoort et al. (2011). The juices were clarified, centrifuged and filtered analogous to the organic acids analysis (Section 2.2.2). A dilution (1/10) of the filtrate in milli-Q water was made prior to analysis in RP-HPLC system with evaporative light scattering detection (Alltech 3300 ELSD, Grace, Deerfield, IL). The injection volume was 5 μL. Separation was performed on a Prevail carbohydrate ES column (250 mm × 4.6 mm, 5 μm particle size, Alltech Grace, Deerfield, IL) coupled to a Prevail C_{18} guard cartridge. Isocratic elution (75% (v/v) acetonitrile/water) was applied at a flow rate of 1 mL min^{-1} at 30 °C. Sugar analyses were carried out in triplicate. Identification was performed by