Food Research International 54 (2013) 169–176

Contents lists available at ScienceDirect

Food Research International

journal homepage: www.elsevier.com/locate/foodres

# Effect of high pressure homogenization (HPH) on the rheological properties of tomato juice: Creep and recovery behaviours

Pedro E.D. Augusto [a,b,*], Albert Ibarz [c], Marcelo Cristianini [a]

[a] Department of Food Technology (DTA), School of Food Engineering (FEA), University of Campinas (UNICAMP), Brazil
[b] Technical School of Campinas (COTUCA), University of Campinas (UNICAMP), Brazil
[c] Department of Food Technology (DTA), School of Agricultural and Forestry Engineering (ETSEA), University of Lleida (UdL), Lleida, Spain

ARTICLE INFO

*Article history:*
Received 7 February 2013
Accepted 26 June 2013

*Keywords:*
Creep–recovery characterization
Compliance
High pressure homogenization
Rheology
Tomato juice

ABSTRACT

High pressure homogenization (HPH) is a non-thermal technology that has been proposed as a partial or total substitute for the thermal processing of food. Although its effect on microbial inactivation has been widely studied, the rheological changes occurring in fruit products need better describing. The present work evaluated the effect of HPH (up to 150 MPa) on the creep and recovery properties of tomato juice. The mechanical Burger's model explained juice creep compliance well, and its parameters (Newtonian dashpots and Hookean springs) were evaluated as a function of the homogenization pressure. HPH processing increased both tomato juice elastic and viscous behaviours, which could be attributed to the disruption of suspended particles during processing. Moreover, each Burger's model constituent could be related to the product internal structure. The results obtained highlighted the possible applications of the HPH process as a valuable tool to promote physical changes in food products.



## 1. Introduction

The rheological characterization of food is important for the design of unit operations, process optimization and high quality product assurance (Ibarz & Barbosa-Cánovas, 2003; Rao, 1999). The viscoelastic properties are very useful in the design and prediction of product stability (Ibarz & Barbosa-Cánovas, 2003), which is important in the description of many processing and storage phenomena (Steffe, 1996).

The high pressure homogenization (HPH) technology consists of pressurizing a fluid to flow through a narrow gap valve, which greatly increases its velocity, resulting in depressurization with consequent cavitation and high shear stress. Thus the macromolecules and suspended particles in the fluid (as cells and its fragments) are subjected to high mechanical stress, which are twisted and deformed (Floury, Bellettre, Legrand, & Desrumaux, 2004; Pinho, Franchi, Augusto, & Cristianini, 2011). This technology has been studied by many authors as a non-thermal food preservation technique, especially for fruit and vegetable products, such as for tomato (Corbo, Bevilacqua, Campaniello, Ciccarone, & Sinigaglia, 2010), apple (Donsì, Esposito, Lenza, Senatore, & Ferrari, 2009; Pathanibul, Taylor, Davidson, & Harte, 2009; Saldo, Suárez-Jacobo, Gervilla, Guamis, & Roig-Sagués, 2009), mango (Tribst, Franchi, Cristianini, & Massaguer, 2009, 2011), orange (Campos & Cristianini, 2007), carrot (Pathanibul et al., 2009; Patrignani, Vannini, Kamdem, Lanciotti, & Guerzoni, 2009, 2010), banana (Calligaris, Foschia, Bastolomeoli, Maifreni, & Manzocco, 2012) and apricot (Patrignani et al., 2009, 2010) juices.

Tomato is one of the most important vegetables in the food industry, which is widely included in the human diet. Homogenization is a commonly used unit operation in tomato processing, and it is well known that homogenization increases the apparent viscosity of tomato products (Bayod, Månsson, Innings, Bergenståhl, & Tornberg, 2007; Bayod & Tornberg, 2011; Beresovsky, Kopelman, & Mizrahi, 1995; Foda & Mccollum, 1970; Lopez-Sanchez, Nijsse, et al., 2011; Mohr, 1987; Ouden & Vliet, 1997, 2002; Thakur, Singh, & Handa, 1995; Whittenberg & Nutting, 1958). However, there are only a few works related to the effect of high pressure homogenization (HPH) on tomato product rheology, especially with respect to the viscoelastic properties.

Augusto, Ibarz, and Cristianini (2012b) evaluated the effect of HPH (up to 150 MPa) on the time-dependent and steady-state shear rheological properties of tomato juice. Augusto, Ibarz, and Cristianini (2013) have also studied the effect of HPH on the storage modulus ($G'$), loss modulus ($G''$) and complex viscosity ($\eta^*$) (parameters related to the viscoelastic properties) of tomato juice, as well as the applicability of the Cox–Merz rule was evaluated. However, there is limited number of works in the literature using creep and recovery procedures in order to evaluate vegetable products, in special tomato juice.

Ibarz and Barbosa-Cánovas (2003) observed that by carrying out creep and recovery experiments, it is possible to describe the rheological behaviour of the product using mechanical models and

* Corresponding author at: COTUCA/UNICAMP, R. Culto à Ciência, 177, Botafogo, CEP, 13020-060, Campinas, SP, Brazil. Tel.: +55 19 3521 9950.
*E-mail address:* pedro@cotuca.unicamp.br (P.E.D. Augusto).

0963-9969/$ – see front matter 
http://dx.doi.org/10.1016/j.foodres.2013.06.027

constitutive equations, combining Newton's viscosity equation and Hooke's elasticity equation.

The creep and recovery experiments have been successfully used for the rheological evaluation of food products. The effect of different ingredient addition and processing conditions on the rheology of food products was evaluated in emulsions (Dolz, Hernández, & Delegido, 2008; Yilmaz, Karaman, Dogan, Yetim, & Kayacier, 2012), organogels (Toro-Vazquez, Morales-Rueda, Malia, & Weiss, 2010), salad dressing (Zhang, Quek, Lam, & Easteal, 2008), ketchup-processed cheese mixtures (Karaman, Yilmaz, Cankurt, Kayacier, & Sagdic, 2012), milk (Bayarri, Dolz, & Hernández, 2009), wheat dough (Bockstaele, Leyn, Eeckhout, & Dewettinck, 2011), lentil starch (Ahmed & Auras, 2011) and ice cream (Shama & Sherman, 1966; Sherman, 1966). By conducting creep and recovery experiments, Shama and Sherman (1966) and Sherman (1966) not only evaluated the rheological properties of ice cream mix and thawed ice cream, but could also identify and correlate each Kelvin–Voigt model parameter (see Fig. 1 and Eq. (2)) to the internal structures of the products. Therefore, the creep and recovery experiments were proven to be a valuable tool in order to characterize the viscoelastic properties of some food materials.

The present work evaluated the effect of high pressure homogenization (HPH) on the viscoelastic properties of tomato juice as related to its behaviour in the creep and recovery tests.

## 2. Materials and methods

As described by Augusto, Ibarz, and Cristianini (2012b), a 4.5°Brix tomato juice was obtained by diluting a commercial 30°Brix pulp in distilled water. The commercial pulp was used to guarantee standardization and repeatability. It was obtained using the hot break process, concentrated by evaporation at 65 °C, thermally processed by the UHT method and aseptically packaged.

The pulp was fractionated into small portions in the laboratory, packaged in high density polyethylene bottles and frozen at −18 °C. This procedure allowed for the use of the same product throughout the whole project. Before use, the samples were thawed at 4 °C and diluted using distilled water at 50 °C to ensure better hydration (Tehrani & Ghandi, 2007). The juice was then allowed to rest for 24 h at 5 °C to ensure complete hydration and the release of incorporated air. The pH of juice was 4.6.

### *2.1. The high pressure homogenization (HPH) process*

The juice was homogenized at 0 MPa (control), 25 MPa, 50 MPa, 100 MPa and 150 MPa using a high pressure homogenizer (Panda Plus, GEA Niro Soavi, Italy). The samples were placed into the equipment at room temperature, and quickly cooled in an ice bath placed just after the homogenization. The maximum temperature reached was ~40 °C, for the sample of 150 MPa just before the ice bath. The experiments were carried out with four replicates.

According to Hayes, Smith, and Morris (1998), the tomato products are generally homogenized at 6–10 MPa, homogenization pressures smaller than those evaluated in the present work. However, the expected behaviour of the traditional homogenization process can be evaluated by observing the obtained behaviour and differences between the evaluated homogenization pressures.

### *2.2. Rheological procedures*

Rheological analyses were carried out using a controlled stress ($\sigma$) rheometer (AR2000ex, TA Instruments, USA) and a four-blade vane geometry (bob diameter = 28 mm; cup diameter = 30 mm; bob length = 42 mm; bottom vertical gap = 4.0 mm). The temperature was maintained constant at 25 °C using a Peltier system.

Previously, oscillatory stress sweeps between 0.01 and 10 Pa were carried out at a frequency of 1 Hz to determine the linear viscoelastic region (LVR) of the samples. The LVR limit of the 0 MPa tomato juice was set at 0.1 Pa (Augusto, Falguera, Cristianini, & Ibarz, 2013; Augusto, Ibarz, & Cristianini, 2013), while the limits of the other samples (25–150 MPa) were close to 1.0 Pa (Augusto, Ibarz, & Cristianini, 2013). Thus, a shear stress of 0.1 Pa was selected for the creep procedure, since it could be used for all the samples. Therefore, all the experiments were carried out at the sample LVR.

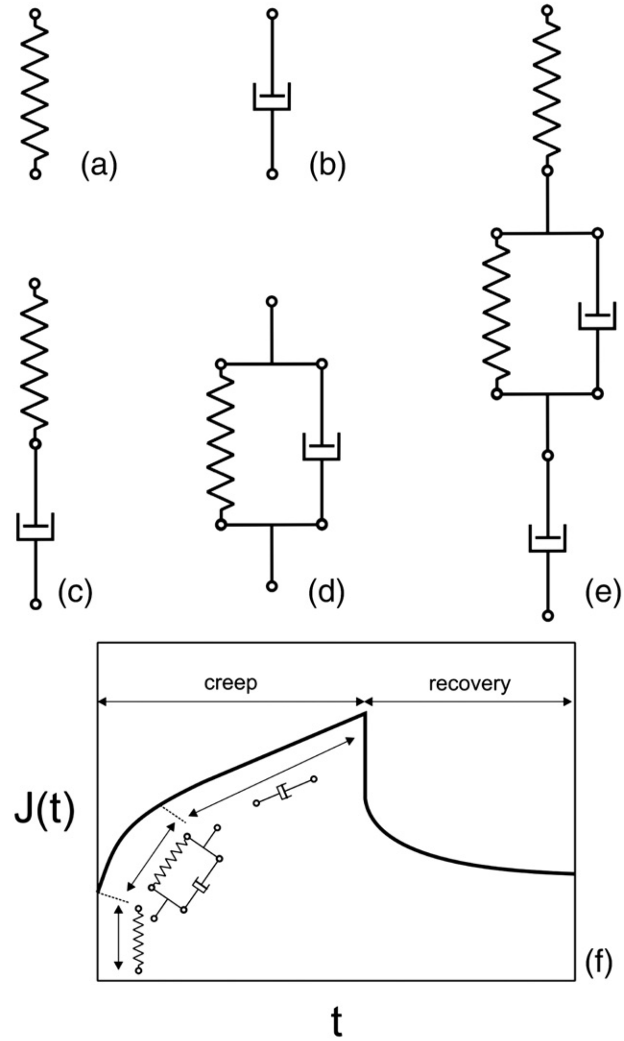


**Fig. 1.** Mechanical models used to describe the viscoelastic properties of foods: (a) the Hookean spring, (b) the Newtonian dashpot, (c) the Maxwell model, (d) the Kelvin–Voigt model, (e) the Burger's model, and (f) a typical creep and recovery plot described by the Burger's model.
Adapted from Ibarz and Barbosa-Cánovas (2003), Rao (1999) and Steffe (1996).

The samples were then placed in the rheometer plate and left to rest for 10 min before the experiment. The creep procedure was carried out at 0.1 Pa for 5 min, and the sample strain ($\gamma$) was recorded. The stress was then removed ($\sigma_{applied}$ = 0.0 Pa), and the sample strain ($\gamma$) recorded for a further 5 min during the recovery procedure. The analyses were conducted with two replicates.

### *2.3. Rheological property evaluation*

The results were expressed according to the compliance function against time (J(t); Eq. (1)). The creep and recovery portions were then evaluated.

$$J(t) = \frac{\gamma(t)}{\sigma_{applied}} \quad (1)$$

The creep behaviour of the tomato juice was described by the Burger's model (Eq. (2)). The Burger's model comprises a Kelvin–Voigt model (a Hookean spring and a Newtonian dashpot placed in parallel) and a Maxwell model (a Hookean spring and a Newtonian dashpot placed in series) placed in series, as illustrated in Fig. 1. It has been widely used to describe the creep behaviour of different biological materials, such as ketchup-processed cheese mixtures (Karaman et al., 2012), lentil starch (Ahmed & Auras, 2011), wheat dough (Bockstaele et al., 2011), organogels (Toro-Vazquez et al., 2010), emulsions (Dolz et al., 2008; Yilmaz et al., 2012) and frozen and melted ice cream (Sherman, 1966).

$$J(t) = \frac{1}{G_0} + \frac{1}{G_1}\left(1 - \exp\left(\frac{-G_1 t}{\eta_1}\right)\right) + \frac{t}{\eta_0} \tag{2}$$

According to Dolz et al. (2008), theoretically, the complete system regeneration will only occur at an infinite time, and will only be complete in the case of a purely elastic system; thus, it is expected that the sample recovery behaviour deviates from the creep behaviour. Therefore, compliance during the recovery procedure was described by an exponential decay function (Eq. (3)), similar to that evaluated by Karaman et al. (2012), Yilmaz et al. (2012), Toro-Vazquez et al. (2010), Bayarri et al. (2009) and Dolz et al. (2008).

$$J(t) = J_\infty + J_{KV} \cdot \exp\left(-B \cdot t^C\right) \tag{3}$$

The parameters of each model were obtained by non-linear regression using the CurveExpert Professional software v.1.2.3, using a significant probability level of 95%. Moreover, the effect of homogenization pressure ($P_H$) on the parameters of Eqs. (2) and (3) was evaluated using the analysis of variance (ANOVA) and the Tukey test at a 95% confidence level. The STATISTICA 5.5 (StatSoft, Inc., USA) software was used for this purpose.

### 2.4. Particle size distribution (PSD) and microstructure

In order to better explain the obtained rheological results, some results previously published by Augusto, Ibarz, and Cristianini (2012b) and Kubo, Augusto, and Cristianini (2013) were included, relating to the suspended particle characterization.

The sample particle size distribution (PSD) was measured by light scattering. The results are reported by Augusto, Ibarz, and Cristianini (2012b) and Kubo et al. (2013). The volume-based mean diameter (D[4,3], Eq. (4), where $n_i$ is the number of particles with diameter $d_i$) and the area-based mean diameter (D[3,2], Eq. (5)) were also evaluated. Both properties were evaluated since the D[4,3] is highly influenced by large particles, whereas D[3,2] is more influenced by the smaller ones (Bengtsson & Tornberg, 2011; Lopez-Sanchez, Nijsse, et al., 2011).

$$D[4,3] = \frac{\sum_i n_i d_i^4}{\sum_i n_i d_i^3} \tag{4}$$

$$D[3,2] = \frac{\sum_i n_i d_i^3}{\sum_i n_i d_i^2} \tag{5}$$

The juice microstructure was evaluated using optical microscopy. The results are reported by Kubo et al. (2013).

## 3. Results and discussion

### 3.1. Creep and recovery behaviours and microstructure evaluation

Fig. 2 shows the creep and recovery behaviours of the tomato juice as a function of the homogenization pressure ($P_H$). As expected, the increase in $P_H$ reduced juice compliance due to the development of a stronger internal structure. This behaviour was in agreement with our previous results for steady-state shear and viscoelasticity (Augusto, Ibarz & Cristianini, 2012b, 2013), and also with those obtained by Bayod and Tornberg (2011), Lopez-Sanchez, Nijsse, et al. (2011), Bayod et al. (2007) and Ouden and Vliet (2002, 1997).

Fig. 2 also shows that the effect of HPH followed an asymptotic behaviour in relation to the $P_H$, i.e., the product changes were higher at "low" $P_H$ values, and progressively lower at "high" $P_H$ values. This is also in agreement with the results previously observed for the steady-state shear and viscoelastic behaviour (Augusto, Ibarz & Cristianini, 2012b, 2013), as well as for the PSD.

Augusto, Ibarz, and Cristianini (2012b) studied the effect of HPH (up to 150 MPa) on the time-dependent and steady-state shear rheological properties of tomato juice. The HPH processing reduced both the tomato juice mean particle diameter and particle size distribution (PSD), increasing the particle surface area and the interaction forces among them. Consequently, HPH increased the juice consistency and thixotropy. In fact, Bengtsson and Tornberg (2011), Lopez-Sanchez, Nijsse, et al. (2011), Bayod, Willers, and Tornberg (2008) and Ouden and Vliet (1997) observed that the yield stress ($\sigma_0$) of tomato products increased due to homogenization ($P_H$ < 60 MPa).

Augusto, Ibarz, and Cristianini (2013) studied the effect of HPH (up to 150 MPa) on the viscoelastic properties of tomato juice. HPH increased the tomato juice storage (G′) and loss (G″) moduli, increasing both the elastic and viscous behaviours of the product. Moreover, HPH processing improved the consistency of the tomato juice more than it modified its nature/behaviour (i.e., changes on the consistency coefficients – viscoelastic property power law model – were higher than those on the behaviour index). The changes observed in the viscoelastic properties were attributed to disruption of the suspended particles during processing. Similar results were observed by Ouden and Vliet (2002), Bengtsson and Tornberg (2011) and Bayod and Tornberg (2011).

Fig. 3 shows that increasing the $P_H$ progressively reduced the diameter of the juice suspended particles. Moreover, the changes in particle diameter between 50 MPa and 150 MPa were less pronounced than those between 0 MPa and 50 MPa. Thus the effect of homogenization pressure ($P_H$) on the disruption of suspended particles seems to follow an asymptotic behaviour, i.e., increases in $P_H$ have reduced effects at higher $P_H$ values. Further, this can be observed

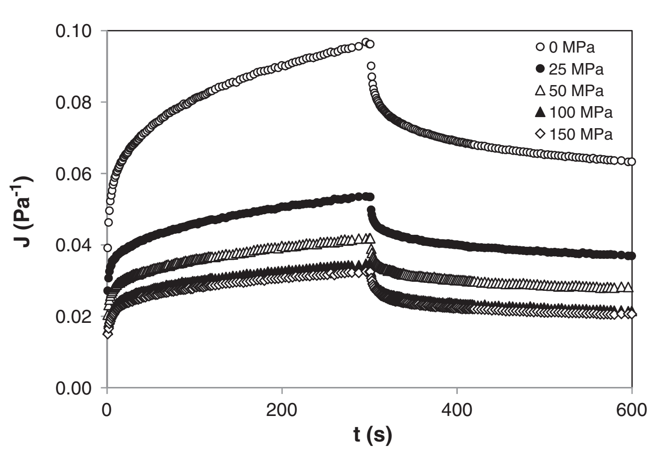


**Fig. 2.** Tomato juice creep and recovery behaviours: effect of HPH.

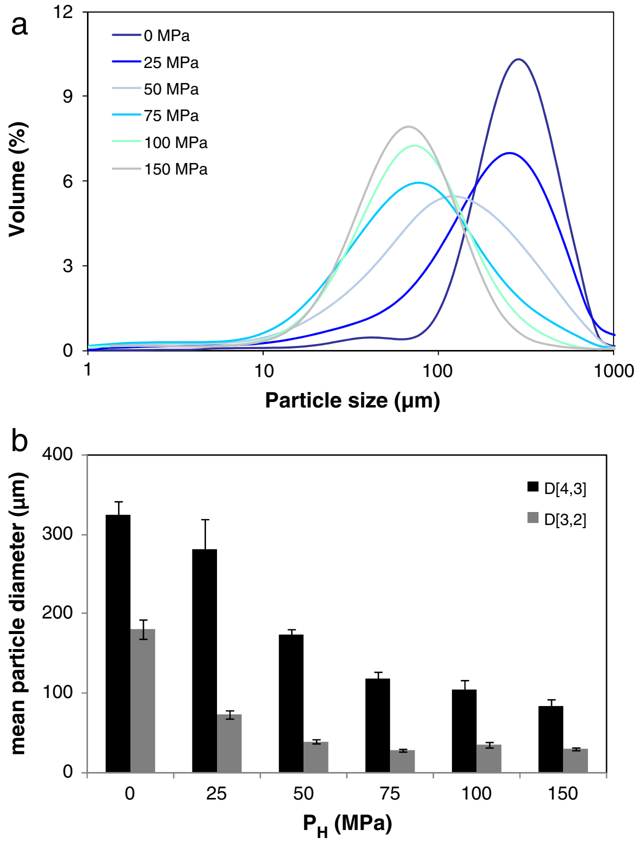


Fig. 3. Effect of HPH on the tomato juice particle size distribution (PSD — a) and particle mean diameter (D[4,3] and D[3,2] — b). Data from Augusto, Ibarz, and Cristianini (2012b) and Kubo et al. (2013).

even in the D[4,3] and D[3,2] values (Fig. 3b), where the reduction in D[3,2] between 0 MPa and 50 MPa (~79%) was higher than in D[4,3] (~45%), indicating a considerable increase in the number of small particles. On the other hand, the reduction in D[3,2] between 50 MPa and 150 MPa (~20%) was smaller than in D[4,3] (~53%), which indicates that the following disruptions occurred preferentially in the larger suspended particles, in accordance with the PSD values shown in Fig. 3a. In conclusion, the increasing $P_H$ progressively reduced the diameter of the larger suspended particles, while that of the smaller particles remained quasi-constant after an initial reduction close to 50 MPa (Fig. 3).

Fig. 4 shows the tomato juice microstructure by optical microscopy. Whereas the control samples showed whole cells, with intact membranes and the characteristic lycopene crystals, the homogenized samples just showed a large amount of small particles, composed of cell walls and internal constituents suspended in the juice serum (Fig. 4). As expected and confirmed by the PSD analysis, the suspended particles were smaller at higher $P_H$ values, highlighting the effect of HPH in disrupting the fruit pulp particles.

The homogenization process disrupts the remaining suspended cells and breaks their fragments into small suspended particles. One would expect the smaller fragments to be less susceptible to breakage during processing when compared to the larger ones or even to the entire cells. Consequently, as the $P_H$ increased, so the larger particles were broken more and more, while the smaller ones remained intact, explaining the PSD behaviour observed. Therefore, the asymptotic behaviour of the rheological properties in relation to the $P_H$ was attributed to the disruption behaviour of suspended particles due to the HPH.

Moelants et al. (2013) described a different disruption behaviour in relation to small and large particles of carrot. In addition to the different behaviours of tomato and carrot particles due to shearing

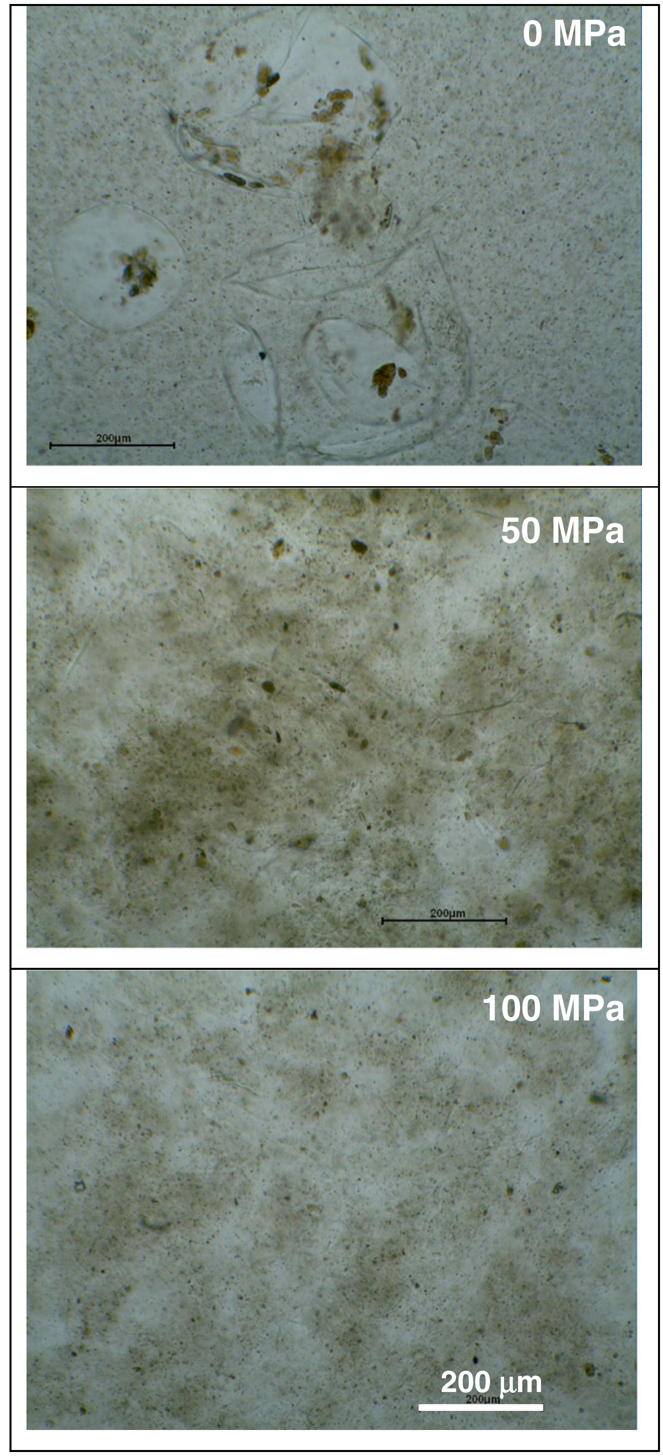


Fig. 4. Effect of HPH on the tomato juice microstructure: optical microscopy. The scale bar shows 200 µm.
Data from Kubo et al. (2013).

(Bengtsson & Tornberg, 2011; Lopez-Sanchez, Svelander, Bialek, Schummm, & Langton, 2011), this difference can be attributed to the sample structure. Moelants et al. (2013) observed that the cell breakage occurred during mechanical breakup rather than cell separation along the middle lamella. In the present work, however, the initial juice showed the presence of whole cells but not of tomato tissues, reinforcing the initial proposition (Fig. 4).

Moreover, the small particles resulting from the higher $P_H$ values tended to form aggregates. In fact, it can be verified by the increase on tomato juice thixotropy (Augusto, Ibarz, & Cristianini, 2012b), directly describing the changes on juice microstructure. Further, it can be seen in the microscopy of Fig. 4. As explained by Augusto, Ibarz, and Cristianini (2012b), cell disruption and subsequent fragmentation not only increased the surface area of the suspended particles, but also changed the properties of the particles and serum. Cell fragmentation exposed and released wall constituents such as pectins and proteins, improving the particle–particle interactions and resulting in aggregates.

### 3.2. Mechanical modeling: creep behaviour

Fig. 5 shows the effect of HPH on the mechanical Burger's model parameters. The creep compliance behaviour of the tomato juice could be well described by the Burger's model (Eq. (2)), with a high coefficient of determination ($R^2 > 0.98$). In Burger's model, $G_0$ is the instantaneous elastic modulus, associated with the Maxwell spring, and $G_1$ is the retarded elastic modulus, associated with the Kelvin–Voigt spring. Moreover, $\eta_0$ is the Newtonian flow viscosity, associated with the Maxwell dashpot, and $\eta_1$ is the retarded viscosity, associated with the Kelvin–Voigt dashpot.

The creep compliance behaviour can be described by models combining larger numbers for the Newtonian ($\eta_n$) and Hookean ($G_n$) elements, such as the six-element model used by Yoo and Rao (1996) to describe the behaviour of tomato concentrates (21–32°Brix). However, especially from an engineering standpoint, a model becomes better, the less the number of associated parameters. Thus the four-element Burger's model was considered adequate to describe the tomato juice creep compliance behaviour.

As the $P_H$ was increased, a gradual increase in $G_0$ and $\eta_0$ was observed. However, the values for $G_1$ and $\eta_1$ showed the same asymptotic behaviour previously observed for $G'$ and $G''$ (Augusto, Ibarz & Cristianini, 2013) and for k, n and $\sigma_0$ (Augusto, Ibarz, & Cristianini, 2012b), with greater changes at $P_H < 25$ MPa and constant values at higher $P_H$ values.

Furthermore, the increase in $P_H$ progressively reduced the diameter of the larger suspended particles, while the smaller particles remained quasi-constant (Augusto, Ibarz, & Cristianini, 2012b). Therefore, the $G_0$ and $\eta_0$ values are mainly affected, and thus could be mainly associated, with the behaviour of the larger particles, whereas the $G_1$ and $\eta_1$ values are mainly affected, and thus could be mainly associated, with the smaller ones (Figs. 3b and 5).

Fruit juices are composed of an insoluble phase (the pulp) dispersed in a viscous solution (the serum). The dispersed phase or pulp is formed of fruit tissue cells and their fragments (cell walls and organelles), and insoluble polymer clusters and chains. The serum is an aqueous solution of soluble polysaccharides, sugars, salts and acids. Thus the rheological properties of the fruit juices are defined by the interactions inside each phase and between them (Augusto, Ibarz, & Cristianini, 2012a). The tomato juice serum is a Newtonian fluid, which becomes a Herschel–Bulkley fluid due to the dispersion of the pulp particles (Augusto, Falguera, Cristianini, & Ibarz, 2012; Augusto, Ibarz, & Cristianini, 2012a; Tanglertpaibul & Rao, 1987).

Cell disruption and further fragmentation during HPH not only increase the surface area of the suspended particles, but also change the particle and serum properties. Cell fragmentation exposes and releases wall constituents, such as pectins and proteins, increasing particle–particle and particle–serum interactions (Augusto, Ibarz, & Cristianini, 2012b). Although a repulsion behaviour between these structures is possible, all the obtained results for tomato juice showed an increase in the attractive interactions, which can be observed not only by the creep behaviour and microstructure results (Figs. 2 and 4), but also by the thixotropic, flow, elastic and viscous results (Augusto, Ibarz & Cristianini, 2012b, 2013).

When the tomato serum is evaluated separately, a small reduction in viscosity is observed due to HPH (~15% at 150 MPa; Augusto, Ibarz, & Cristianini, 2012a). However, when cell fragmentation is considered, the cell constituent release into the serum can increase its viscosity. It is important to highlight that concentration increases the viscosity and consistency exponentially (Ibarz & Barbosa-Cánovas, 2003; Rao, 1999; Steffe, 1996), as can be seen by varying concentrations of cell wall constituents (Augusto, Falguera, Cristianini, & Ibarz, 2011).

The Hookean (G) elements describe the elastic behaviour of a material, while the Newtonian ($\eta$) elements describe its viscous behaviour. Thus, the Hookean (G) elements can be associated with the interactions among the suspended particles (particle–particle and aggregate–aggregate interactions), while the Newtonian ($\eta$) elements can be associated both with the serum behaviour (i.e., the resistance of the fluid to flow; to be deformed) and with the interaction among the particles and serum (i.e., due to the particle drag, also related to the fluid resistance to flow). This explains why the suspended particles can be associated with a Newtonian behaviour, related to the Burger's dashpots ($\eta_0$, $\eta_1$).

The results here obtained, associated with the previous ones (Augusto, Ibarz, & Cristianini, 2012a,b, 2013) permit the following interpretation of the effect of HPH processing on the structure and rheological behaviour of tomato juice.

As the $P_H$ is increased, so the diameter of the larger suspended particles was progressively reduced, while the diameter of the smaller particles was greatly reduced at “low” $P_H$ (<50 MPa), and then remained quasi-constant (Fig. 3b). As the particles were disrupted, so their components were passed to the serum, increasing its viscosity. Moreover, the surface area of the particles increased, resulting in greater inter-particle (i.e., elastic) and particle–serum (i.e., viscous) interactions. The reduction in particle diameter increases the surface area, resulting in greater interaction among the particles. Thus, it

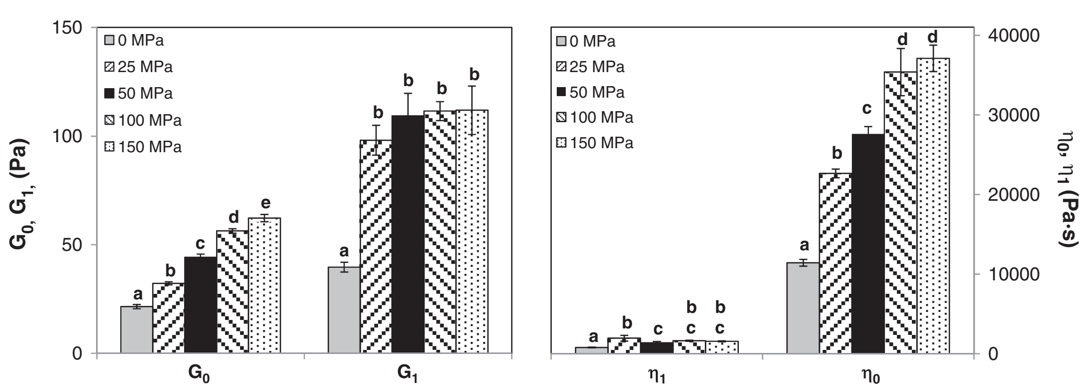


**Fig. 5.** Tomato juice creep compliance behaviour: effect of HPH on the mechanical Burger's model parameters. Mean value at 25 °C; vertical bars represent the standard deviation in each value. For each parameter, different letters represent significantly different values ($P < 0.05$).

increases the elastic behaviour ($G_0$, $G_1$) of the particles. Moreover, the release of soluble materials from the particles and the formation of aggregates increase the viscous behaviour ($\eta_0$, $\eta_1$) of the product, both, due to the particle drag and also to the fluid resistance to flow. Since the major particle changes are associated with a $P_H$ of up to 50 MPa (Fig. 3), the main rheological changes take place at this processing level (Fig. 2).

Non-hydrodynamic forces can be important in systems with smaller suspended particles, such as the HPH tomato juice (Augusto, Ibarz, & Cristianini, 2012b). Small particle interaction can be due to van der Waals forces (Genovese, Lozano, & Rao, 2007; Tsai & Zammouri, 1988) and/or electrostatic forces due to the interaction between the negatively charged pectins and the positively charged proteins (Beresovsky et al., 1995; Takada & Nelson, 1983). Thus, it is expected that HPH processing would result in small particle aggregation with the formation of a network, as described by Bayod and Tornberg (2011), resulting in a more thixotropic fluid (Augusto, Ibarz, & Cristianini, 2012b). Hence, as the $P_H$ is increased, so a large number of aggregates are expected.

### *3.3. Correlating the juice rheological behaviour with the Burger's mechanical model*

Vegetable juices are very complex systems, containing many different components whose interactions are complex and difficult to predict. Even so, trying to describe and modeling their behaviour are interesting by a scientific and an engineering standpoint. When it is successfully attained, the obtained model is a simplification of the real condition, with limitations and even empirical or speculative propositions.

Therefore, considering the described creep behaviour, compared with the PSD, microstructure and previous rheological (especially the thixotropic ones) results, as well as the theoretical response of each element, the elements of the Burger's model could be mainly associated with each tomato juice component.

Considering that the juice is composed by three elements: isolated particles, particle aggregates and serum.

The particles and aggregates are dispersed through the serum, and when they are moving, a Newtonian (viscous) response is observed due to drag. Thus, the Burger's Newtonian components can be attributed both to particles (isolated and forming aggregates) and serum.

The Burger's Hookean elements, however, can only be attributed to the particles. The particle interactions result in an elastic (Hookean) response, either inside the aggregates (i.e., the particles that are forming the aggregates) or isolated.

The Burger's model comprises a Kelvin–Voigt and a Maxwell component placed in series (Fig. 1).

The Kelvin–Voigt element is composed of a Hookean spring and a Newtonian dashpot placed in parallel (Fig. 1). Thus, both elastic and viscous responses take place simultaneously (it can be better understood in Fig. 1f). It suggests that the particle aggregates are the main elements related to the observed Kelvin–Voigt element, as both elastic (particle interactions) and viscous (drag through the serum) behaviours are present during flow.

The Maxwell element is composed of a Hookean spring and a Newtonian dashpot placed in series (Fig. 1). Thus, the elastic and viscous responses take place at different moments (it can be better understood in Fig. 1f). When the element is subjected to shear, the elastic response is firstly observed, which is followed by the viscous one. Thus, it suggests that when the juice is submitted to flow, the suspended particles, which are dispersed and spaced, show an elastic behaviour, due to the tendency of being approximated (similar to the behaviour of a spring that is suddenly caught by a runner). Then, after reaching the equilibrium, those particles flow through the serum, whose drag result in the fluid Newtonian response.

Following these assumptions, each model element can be mainly correlated with a juice internal component.

The first Hookean element ($G_0$), associated with the Maxwell spring, describes the elastic behaviour of the product and could be mainly related to the behaviour of the large particles during HPH (i.e., the D[4,3] results — Fig. 3b). Thus it could be mainly associated with the suspended particles, i.e., to the inter-particle interactions.

The first Newtonian element ($\eta_0$), associated with the Maxwell dashpot, describes the viscous behaviour of the product and could also be related to the behaviour of the larger particles during HPH (Fig. 3b). Therefore it can be mainly associated with the resistance of the product to flow, which related to serum viscosity and to the contribution of particle drag during flow (i.e., particle–serum interaction).

The Kelvin–Voigt element could be mainly related to the behaviour of the smaller particles during HPH (i.e., the D[3,2] results — Fig. 3b), and the small particles were related to particle aggregate formation. Therefore, the Kelvin–Voigt element can be associated with the formation of aggregates by the tomato juice particles. Moreover, the Kelvin–Voigt Hookean element ($G_1$) can be mainly associated with inter-particle interactions inside the aggregates and between aggregates, while the Kelvin–Voigt Newtonian element ($\eta_1$) can be mainly associated with aggregate drag during flow (i.e., aggregate–serum interactions).

Therefore a better understanding of the rheological behaviour of the tomato juice, as well as the influence of HPH, could be obtained by using the creep compliance analysis. Fig. 6 shows the proposed description of the tomato juice, obtained using the Burger's model.

Once more we highlighted that the proposed interpretation is not a complete and final model for the tomato juice rheology. Other neglected interactions can also be important. In fact, it is almost impossible to prove most of the proposed interactions. However, even containing empirical or speculative propositions, we believe that it can be considered a simplification of the tomato juice rheological behaviour, which is interesting by either a scientific or an engineering standpoint.

In fact, we also believe that new models should contain some empirical or speculative propositions, always based on solid scientific information and knowledge, in order to try to simplify and then describe the complex world around us.

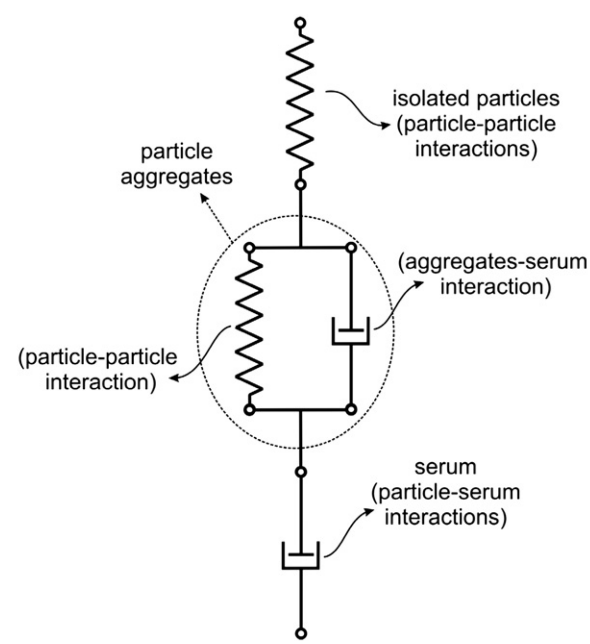


**Fig. 6.** Proposed model to describe the tomato juice creep compliance behaviour in relation to the observed main interactions.

### 3.4. Mechanical modeling: recovery behaviour

After the creep period, the applied stress was immediately removed ($\sigma_{applied} = 0.0$ Pa), and sample compliance (J) recorded during the recovery procedure. The compliance behaviour of the tomato juice during recovery can be well described by the exponential decay model (Eq. (3)), with a high coefficient of determination ($R^2 > 0.97$). Fig. 7 shows the effect of HPH on the compliance behaviour of the tomato juice during recovery, considering the exponential decay model described (Eq. (3)). The parameters B and C are the proportional and power kinetic parameters, respectively, in the recovery model. Thus, the parameter B is related to the magnitude of the time involved in compliance decay, while C describes the curvature shape. The compliance $J_\infty$ is the residual compliance corresponding to the permanent deformation of the Maxwell dashpot, while $J_{KV}$ is the recovery compliance due to the Kelvin–Voigt element.

Both compliance parameters ($J_\infty$, $J_{KV}$) showed an asymptotic-decreasing behaviour in relation to the $P_H$ (i.e., increasing the resistance of deformation), again indicating a strengthening of the internal structure of the tomato juice due to HPH. The compliance $J_\infty$ is related to the Maxwell dashpot and to permanent sample deformation (i.e., the energy dissipated to flow). As expected (since $J_\infty = t / \eta_0$; Eq. (2)), its behaviour was similar to that obtained by the first Newtonian element ($\eta_0$, Fig. 5). Furthermore, the compliance $J_{KV}$ is the recovery compliance due to the Kelvin–Voigt element, which is a function of $G_1$ and $\eta_1$ (Eq. (2)). Once again, its behaviour was similar to that obtained by the Kelvin–Voigt spring and dashpot (Fig. 5). The proportional kinetic parameter (B) showed no dependence on the $P_H$, which is close to 0.25 $s^{-C}$ for all the samples. The power kinetic parameter (C) showed values close to 0.35 at $P_H \leq 50$ MPa, and then a slight increase at $P_H \geq 100$ MPa (~0.4).

Thus using the creep and recovery experiments, it was possible to better understand the effect of high pressure homogenization (HPH) on the rheological properties of tomato juice. The results obtained described the effect of HPH on the internal structure of tomato juice, indicating possible applications for this process as a valuable tool to promote changes in the physical properties of food products, such as improving both their elastic and viscous behaviours. Thus, HPH can be used to increase the consistency of tomato juice, improving its sensory acceptance, reducing the need for adding hydrocolloids and reducing particle sedimentation or serum separation.

## 4. Conclusions

The present work evaluated the effect of high pressure homogenization (HPH) on the creep and recovery properties of tomato juice. The mechanical Burger's model explained the juice creep compliance well, and its parameters were evaluated as a function of the homogenization pressure ($P_H$). HPH processing improved the elastic and viscous behaviours of the tomato juice, and the changes could be related to the internal structure of the product. The results obtained highlighted possible applications of the high pressure homogenization (HPH) process as a valuable tool to promote changes in the physical properties of food products.

*Nomenclature*

| | |
|---|---|
| $\gamma$ | strain [–] |
| $\eta$ | viscosity, associated with the Newtonian dashpot [Pa·s] |
| $\eta_0$ | Newtonian flow viscosity, associated with the Maxwell dashpot [Pa·s] |
| $\eta_1$ | retarded viscosity, associated with the Kelvin–Voigt dashpot [Pa·s] |
| $\eta^*$ | complex viscosity [Pa·s] |
| $\sigma$ | shear stress [Pa] |
| $\sigma_0$ | yield stress, Herschel–Bulkley's model [Pa] |
| B | proportional kinetic parameter in the recovery model (Eq. (3)) [$s^{-C}$] |
| C | power kinetic parameter in the recovery model (Eq. (3)) [–] |
| d | particle diameter [μm] |
| D[4,3] | particle volume-based diameter (Eq. (4)) [μm] |
| D[3,2] | particle area-based diameter (Eq. (5)) [μm] |
| G | elastic modulus, associated with the Hookean spring [Pa] |
| $G_0$ | instantaneous elastic modulus, associated with the Maxwell spring [Pa] |
| $G_1$ | retarded elastic modulus, associated with the Kelvin–Voigt spring [Pa] |
| $G'$ | storage modulus [Pa] |
| $G''$ | loss modulus [Pa] |
| J | compliance (Eq. (1)) [$Pa^{-1}$] |
| $J_\infty$ | residual compliance corresponding to the permanent deformation of the Maxwell dashpot [$Pa^{-1}$] |
| $J_{KV}$ | recovery compliance due to the Kelvin–Voigt element [$Pa^{-1}$] |
| k | consistency coefficient, Herschel–Bulkley's model [$Pa·s^n$] |
| n | flow behaviour index, Herschel–Bulkley's model [–] |
| $P_H$ | homogenization pressure [MPa] |
| t | time [s] |

## Acknowledgments

The authors are grateful to the São Paulo Research Foundation (FAPESP) for funding project nos. 2010/05241-8 and 2010/05240-1.

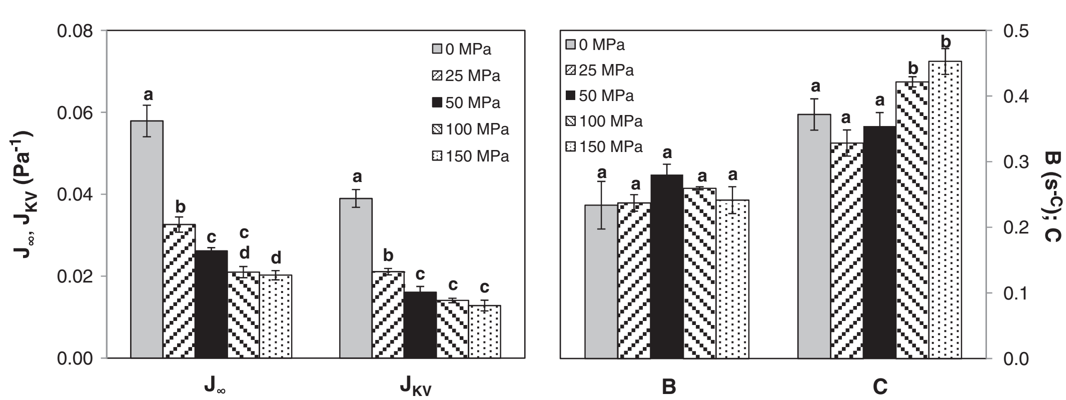


**Fig. 7.** Tomato juice recovery compliance behaviour: effect of HPH on the exponential decay model parameters. Mean value at 25 °C; vertical bars represent the standard deviation for each value. For each parameter, different letters represent significantly different values ($P < 0.05$).

## References


Ahmed, J., & Auras, R. (2011). Effect of acid hydrolysis on rheological and thermal characteristics of lentil starch slurry. *LWT— Food Science and Technology*, *44*, 976–983.

Augusto, P. E. D., Falguera, V., Cristianini, M., & Ibarz, A. (2011). Influence of fibre addition on the rheological properties of peach juice. *International Journal of Food Science and Technology*, *46*, 1086–1092.

Augusto, P. E. D., Falguera, V., Cristianini, M., & Ibarz, A. (2012). Rheological behaviour of tomato juice: Steady-state shear and time-dependent modeling. *Food and Bioprocess Technology*, *5*(5), 1715–1723.

Augusto, P. E. D., Falguera, V., Cristianini, M., & Ibarz, A. (2013). Viscoelastic properties of tomato juice: Applicability of the Cox–Merz Rule. *Food and Bioprocess Technology*, *6*(3), 839–843.

Augusto, P. E. D., Ibarz, A., & Cristianini, M. (2012a). Effect of high pressure homogenization (HPH) on the rheological properties a fruit juice serum model. *Journal of Food Engineering*, *111*, 473–477.

Augusto, P. E. D., Ibarz, A., & Cristianini, M. (2012b). Effect of high pressure homogenization (HPH) on the rheological properties of tomato juice: Time-dependent and steady-state shear. *Journal of Food Engineering*, *111*, 570–579.

Augusto, P. E. D., Ibarz, A., & Cristianini, M. (2013). Effect of high pressure homogenization (HPH) on the rheological properties of tomato juice: Viscoelastic properties and the Cox–Merz rule. *Journal of Food Engineering*, *114*, 57–63.

Bayarri, S., Dolz, M., & Hernández, M. J. (2009). Effect of carboxymethyl cellulose concentration on rheological behaviour of milk and aqueous systems. A creep and recovery study. *Journal of Applied Polymer Science*, *114*, 1626–1632.

Bayod, E., Månsson, P., Innings, F., Bergenståhl, B., & Tornberg, E. (2007). Low shear rheology of concentrated tomato products. Effect of particle size and time. *Food Biophysics*, *2*, 146–157.

Bayod, E., & Tornberg, E. (2011). Microstructure of highly concentrated tomato suspensions on homogenization and subsequent shearing. *Food Research International*, *44*, 755–764.

Bayod, E., Willers, E. P., & Tornberg, E. (2008). Rheological and structural characterization of tomato paste and its influence on the quality of ketchup. *LWT— Food Science and Technology*, *41*, 1289–1300.

Bengtsson, H., & Tornberg, E. (2011). Physicochemical characterization of fruit and vegetable fiber suspensions. I: Effect of homogenization. *Journal of Texture Studies*, *42*, 268–280.

Beresovsky, N., Kopelman, I. J., & Mizrahi, S. (1995). The role of pulp interparticle interaction in determining tomato juice viscosity. *Journal of Food Processing and Preservation*, *19*, 133–146.

Bockstaele, F. V., Leyn, I. D., Eeckhout, M., & Dewettinck, K. (2011). Non-linear creep–recovery measurements as a tool for evaluating the viscoelastic properties of wheat flour dough. *Journal of Food Engineering*, *107*, 50–59.

Calligaris, S., Foschia, M., Bastolomeoli, I., Maifreni, M., & Manzocco, L. (2012). Study on the applicability of high-pressure homogenization for the production of banana juices. *LWT— Food Science and Technology*, *45*, 117–121.

Campos, F., & Cristianini, M. (2007). Inactivation of *Saccharomyces cerevisiae* and *Lactobacillus plantarum* in orange juice using ultra high-pressure homogenization. *Innovative Food Science & Emerging Technologies*, *8*(2), 226–229.

Corbo, M. R., Bevilacqua, A., Campaniello, D., Ciccarone, C., & Sinigaglia, M. (2010). Use of high pressure homogenization as a mean to control the growth of foodborne moulds in tomato juice. *Food Control*, *21*, 1507–1511.

Dolz, M., Hernández, M. J., & Delegido, J. (2008). Creep and recovery experimental investigation of low oil content food emulsions. *Food Hydrocolloids*, *22*, 421–427.

Donsì, F., Esposito, L., Lenza, E., Senatore, B., & Ferrari, G. (2009). Production of self-stable Annurca apple juice with pulp by high pressure homogenization. *International Journal of Food Engineering*, *5*(4) (article 12).

Floury, J., Bellettre, J., Legrand, J., & Desrumaux, A. (2004). Analysis of a new type of high pressure homogeniser. A study of the flow pattern. *Chemical Engineering Science*, *59*, 843–853.

Foda, Y. M., & Mccollum, J. P. (1970). Viscosity as affected by various constituents of tomato juice. *Journal of Food Science*, *35*, 333–338.

Genovese, D. B., Lozano, J. E., & Rao, M. A. (2007). The rheology of colloidal and noncolloidal food dispersions. *Journal of Food Science*, *72*(2), R11–R20.

Hayes, W. A., Smith, P. G., & Morris, A. E. J. (1998). The production and quality of tomato concentrates. *Critical Reviews in Food Science and Nutrition*, *38*(7), 537–564.

Ibarz, A., & Barbosa-Cánovas, G. V. (2003). *Unit operations in food engineering.* Boca Raton: CRC Press.

Karaman, S., Yilmaz, M. T., Cankurt, H., Kayacier, A., & Sagdic, O. (2012). Linear creep and recovery analysis of ketchup-processed cheese mixtures using mechanical simulation models as a function of temperature and concentration. *Food Research International*, *48*, 507–519.

Kubo, M. T. K., Augusto, P. E. D., & Cristianini, M. (2013). Effect of high pressure homogenization (HPH) on the physical stability of tomato juice. *Food Research International*, *51*, 170–179.

Lopez-Sanchez, P., Nijsse, J., Blonk, H. C. G., Bialek, L., Schumm, S., & Langton, M. (2011). Effect of mechanical and thermal treatments on the microstructure and rheological properties of carrot, broccoli and tomato dispersions. *Journal of the Science of Food and Agriculture*, *91*, 207–217.

Lopez-Sanchez, P., Svelander, C., Bialek, L., Schummm, S., & Langton, M. (2011). Rheology and microstructure of carrot and tomato emulsions as a result of high-pressure homogenization conditions. *Journal of Food Science*, *76*(1), E130–E140.

Moelants, K. R. N., Cardinaels, R., Jolie, R. P., Verrijssen, T. A. J., Buggenhout, S. V., Zumalacarregui, L. M., et al. (2013). Relation between particle properties and rheological characteristics of carrot-derived suspensions. *Food and Bioprocess Technology*, *6*(5), 1127–1143.

Mohr, W. P. (1987). Tomato fruit properties affecting consistency of concentrated product. *Canadian Journal of Plant Science*, *67*, 881–890.

Ouden, F. W. C. D., & Vliet, T. V. (1997). Particle size distribution in tomato concentrate and effects on rheological properties. *Journal of Food Science*, *62*(3), 565–567.

Ouden, F. W. C. D., & Vliet, T. V. (2002). Effect of concentration on the rheology and serum separation of tomato suspensions. *Journal of Texture Studies*, *33*, 91–104.

Pathanibul, P., Taylor, T. M., Davidson, P. M., & Harte, F. (2009). Inactivation of *Escherichia coli* and *Listeria innocua* in apple and carrot juices using high pressure homogenization and nisin. *International Journal of Food Microbiology*, *129*, 316–320.

Patrignani, F., Vannini, L., Kamdem, S. L. S., Lanciotti, R., & Guerzoni, M. E. (2009). Effect of high pressure homogenization on *Saccharomyces cerevisiae* inactivation and physic-chemical features in apricot and carrot juices. *International Journal of Food Microbiology*, *136*, 26–31.

Patrignani, F., Vannini, L., Kamdem, S. L. S., Lanciotti, R., & Guerzoni, M. E. (2010). Potentialites of high pressure homogenization to inactivate *Zygosaccharomyces bailii* in fruit juices. *Journal of Food Science*, *75*(2), M116–M120.

Pinho, C. R. G., Franchi, M. A., Augusto, P. E. D., & Cristianini, M. (2011). Milk flow evaluation during high pressure homogenization (HPH) using computational fluid dynamics (CFD). *Brazilian Journal of Food Technology*, *14*(3), 232–240. http://dx.doi.org/10.4260/BJFT2011140300028 (in Portuguese).

Rao, M. A. (1999). *Rheology of fluid and semisolid foods: Principles and applications.* Gaithersburg: Aspen Publishers.

Saldo, J., Suárez-Jacobo, Á., Gervilla, R., Guamis, B., & Roig-Sagués, A. X. (2009). Use of ultra-high-pressure homogenization to preserve apple juice without heat damage. *High Pressure Research*, *29*(1), 52–56.

Shama, F., & Sherman, P. (1966). The texture of ice cream. 2. Rheological properties of frozen ice cream. *Journal of Food Science*, *31*, 699–706.

Sherman, P. (1966). The texture of ice cream. 3: Rheological properties of mix and melted ice cream. *Journal of Food Science*, *31*, 707–716.

Steffe, J. F. (1996). *Rheological methods in food process engineering* (2nd ed.)East Lansing: Freeman Press.

Takada, N., & Nelson, P. E. (1983). Pectin–protein interaction in tomato products. *Journal of Food Science*, *48*(5), 1408–1411.

Tanglertpaibul, T., & Rao, M. A. (1987). Flow properties of tomato concentrates: Effect of serum viscosity and pulp content. *Journal of Food Science*, *52*(2), 318–321.

Tehrani, M. M., & Ghandi, A. (2007). Modification of Bostwick method to determine tomato concentrate consistency. *Journal of Food Engineering*, *79*, 1483–1486.

Thakur, B. R., Singh, R. K., & Handa, A. K. (1995). Effect of homogenization pressure on consistency of tomato juice. *Journal of Food Quality*, *18*, 389–396.

Toro-Vazquez, J. F., Morales-Rueda, J., Malia, V. A., & Weiss, R. G. (2010). Relationship between molecular structure and thermo-mechanical properties of candelilla wax and amides derived from (r)-12-hydroxystearic acid as gelators of safflower oil. *Food Biophysics*, *5*, 193–202.

Tribst, A. A. L., Franchi, M. A., Cristianini, M., & Massaguer, P. R. (2009). Inactivation of *Aspergillus niger* in mango nectar by high-pressure homogenization combined with heat shock. *Journal of Food Science*, *74*(9), M509–M514.

Tribst, A. A. L., Franchi, M. A., Cristianini, M., & Massaguer, P. R. (2011). Quality of mango nectar processed by high-pressure homogenization with optimized heat treatment. *Journal of Food Science*, *76*(2), M106–M110.

Tsai, S. C., & Zammouri, K. (1988). Role of interparticular Van der Waals force in rheology of concentrated suspensions. *Journal of Rheology*, *32*(7), 737–750.

Whittenberg, R. T., & Nutting, G. C. (1958). High viscosity of cell wall suspensions prepared from tomato juice. *Food Technology*, *12*, 420–422.

Yilmaz, M. T., Karaman, S., Dogan, M., Yetim, H., & Kayacier, A. (2012). Characterization of O/W model system meat emulsions using shear creep and creep recovery tests based on mechanical simulation models and their correlation with texture profile analysis (TPA) parameters. *Journal of Food Engineering*, *108*(2), 327–336.

Yoo, B., & Rao, M. A. (1996). Creep and dynamic rheological behaviour of tomato concentrates: Effect of concentration and finisher screen size. *Journal of Texture Studies*, *27*, 451–459.

Zhang, C., Quek, S. Y., Lam, G., & Easteal, A. (2008). The rheological behaviour of low fat soy-based salad dressing. *International Journal of Food Science and Technology*, *43*, 2204–2212.