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Tuning Physical Properties of Tomato Puree by Fortification with Grape Skin Antioxidant Dietary Fibre

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Abstract

Grape skins recovered from winemaking byproducts were investigated for use as sustainable, antioxidant fibre-rich ingredient for the innovation of low energy dense tomato puree. Six tomato purees fortified with grape skin antioxidant fibre, with varying particle size distribution, and two control tomato purees were studied. Physical parameters of purees were analysed upon mixing and either an intensive heat treatment or an optimized heat treatment designed to achieve 6 decimal reduction of a target microorganism (*Alicyclobacillus acidoterrestris*) as recommended for pasteurisation of acidic fruit products.

Mixing of grape skin antioxidant fibre with tomato purees led to a decrease in both surface-weighted mean diameter (Sauter mean diameter, d(3,2)) and volume-weighted mean diameter (d(4,3)) values and an increase in span. Changes in these descriptors were most significant in purees added with the smallest particle sizes. Thermal stabilisation of purees slightly decreased the d(3,2) values further and increased d(4,3) values, suggesting concomitant occurrence of particle disaggregation and formation of flocs within the food matrix. Phenolic solubility was inversely correlated to d(3,2) values. Bostwick consistency, storage (G') and loss (G'') moduli and complex viscosity (η*) increased in the fortified purees. The η* values displayed a positive correlation with d(4,3) values. Variations in Hunter colorimetric parameters were within the acceptability threshold. Overall, the information obtained provides knowledge to assist development of fibre-rich, low energy dense fruit purees.

Keyword Grape skins · tomato puree · total phenolics · colour · rheological properties · particle size distribution

Introduction

Increasing fibre content in foods is a strategy to prevent the occurrence of chronic diseases. In fact, numerous health benefits have been associated with an increased intake of dietary fibre, including reduced risk of coronary heart disease, diabetes, obesity, and some forms of cancer (Mann and Cummings 2009; Perez-Jimenez et al. 2008; Lopez-Oliva et al. 2010). In addition to their health benefits, dietary fibres can provide technological properties to the foods as reviewed by Elleuch et al., 2011. In fact, fibre-rich ingredients were incorporated into food products, including bakery, meat, fish and dairy products as non-caloric bulking agents for partial replacement of other ingredients and also to provide technological properties such as: increased viscosity, ability to form gels and/or emulsions and modify texture. Various fibres, including guar gum, xanthan, tragacanth, pectins, and sodium alginate and chemically modified starches have been studied as thickening agents to process tomato paste into ketchup (Juszczak et al., 2013). Food formulation with fibre-rich ingredient results in changes in overall sensorial properties, and hence fibre addition needs to be tailored according to the consumers' liking (Palzer, 2009; Elleuch et al., 2011).

Winemaking by-products, especially grape skins (GS), are interesting sources of functional dietary fibre because they also contain high amount of antioxidants (Sri Harsha et al., 2013). Hence, these byproducts are referred to as "antioxidant dietary fibre" as they can deliver the physiological effects of both dietary fibre and antioxidants (Saura-Calixto, 1998; Perez-Jimenez et al. 2008). It is worth noticing that in a human study, Perez-Jimenez et al. (2008) have demonstrated that the intake of GS significantly reduces the biomarkers of cardiovascular risk. The use of GS as food ingredients has been investigated to increase the nutritional value and/or to modulate the physical properties of chicken hamburgers (Sáyago-Ayerdi et al. 2009), rye bread (Mildner-Szkudlarz et al., 2011), cheese (Han et al., 2011), ice cream (Sagdic et al., 2012), wheat biscuits (Mildner-Szkudlarz et al., 2013), fish (Riberio et al., 2013), yogurt and salad dressing (Tseng and Zhao, 2013).

Tomato is one of the most important vegetable products and it is mainly consumed as processed products, i.e., pastes, concentrates, ketchup, salsa, etc. Incorporation of GS into a tomato puree could be an interesting novel application for GS, since tomato puree is a "low energy density food", while none of the foods proposed previously for delivering GS is a low caloric food. Hence, formulation of this fibre-rich ingredient with a tomato puree is an appropriate strategy to target the expected health benefit. Nevertheless, there is no knowledge on the effects of addition of grape pomace derived ingredients on fruit purees. The incorporation of GS derived fractions into a semi liquid food, such as tomato puree, requires the design of an effective heat treatment since the presence of spores in GS cannot be ruled out. To achieve pasteurisation of low-pH foods, such as tomato puree, the most heat resistant microorganism among the common spoilage microorganisms found in these foods i.e., *Alicyclobacillus acidoterrestris* has been proposed as a process target. It is a thermoacidophilic, non-pathogenic and sporeforming bacterium (Silva and Gibbs, 2004; Bevilacqua and Corvo, 2011). An optimized continuous heat treatment achieving 6D-reduction of the target microorganism (*A. acidoterrestris*) is considered effective for pasteurisation (Silva and Gibbs, 2004). Microwave treatment has been proposed as an efficient pasteurisation process for fruit juice, due to short processing time (Igual et al., 2014). As microwave heating of puree is very fast, this treatment can mimic an optimized continuous industrial treatment on lab-scale (Page et al., 2012). Intensive autoclave treatments are also common for tomato products (Nisha et al., 2011).

Knowledge on rheological properties of tomato-based products provides important information, as demonstrated by previous studies on tomato microstructure and consistency (Anthon et al. 2010; Bayod & Tornberg, 2011; Juszczak et al., 2013; Moelants et al., 2013; Moelants et al., 2014a). In concentrated tomato suspensions, particle properties of the dispersed phase rather than the serum viscosity dominate the rheological properties (Moelants et al. 2013). Particle size distribution (PSD) of a food suspension affects texture, i.e., perceivable homogeneity and consistency. PSD also affects the rate of release of nutrients and sensory-active compounds from the food matrix during eating. In particular, the volume weighted diameter (d(4,3)) is the most significant parameter in relation to the perceptible texture of the product (Imai et al. 1998), while the surface weighted diameter (d(3,2)) affects the kinetics of release of various compounds from the solid matrix (Walstra 2003).

The aim of the current study was to investigate the physical properties of low energy dense tomato purees added with GS as a healthy ingredient. The particle size of the solids of the tomato-GS purees was varied to obtain six different formulations. The formulations were characterized after mixing and either microwave treatment or autoclave treatment. The information obtained could be of value for the design of fruit purees enriched with dietary fibre.

Material and Methods

Chemicals

The integrated total dietary fibre assay procedure kit was purchased from Megazyme International Ltd (Bray, Ireland). All other chemicals were purchased from Sigma Aldrich Italia (Milan, Italy).

Grape skins

Grape pomace (Chardonnay variety) was kindly provided by a winery located in Northern Italy. At the winery, grapes were pressed with separation of grape solids and must. Then grape stalks were separated with a mechanical destemming and the remaining material was sieved (with a 5 mm sieve) to separate GS from the seeds and frozen to inhibit microbial growth (Lavelli et al, 2006). GS were transported frozen to the lab, dried at 50 °C for about 8 h and finely milled by a rotor mill (Cross Beater Mill, Retsch GmbH, Haan, Germany) at room temperature. The powders obtained were sieved by using the Octagon Digital sieve shaker (Endecotts L.t.d., London, United Kingdom), with three certified sieves (openings: 125, 250 and 500μm), under continuous sieving for 10 min at amplitude 8. Three granulometric fractions were collected, namely: GS-L (250μm < GS-L ≤ 500μm), GS-M

(125µm < GS-M ≤ 250µm) and GS-S (GS-S ≤ 125µm). These fractions were stored under vacuum, in the dark, at 4 °C.

Processing of tomato purees at the industrial plant

Two tomato puree samples, namely PV (smooth puree) and PR (rough puree) were provided by a fruit processing company. At the industrial plant, tomatoes were homogenized and heated to approximately 95 °C by steam injection to inactivate endogenous enzymes (hot-break). The homogenate was then passed hot to through a 0.5 mm-screen (PV) or 1 mm-screen (PR) pulper/finisher to remove seeds and skin fragments and deaerated under vacuum. The finished PV and PR purees were then concentrated at 80 °C under reduced atmospheric pressure to increase dry solids from 5.0% to ~10%, using a tubular heat exchanger and aseptically stored in tank under nitrogen for 6 months before bottling. After bottling, the purees were autoclaved at 115 °C for 5.5 min (Figure 1).

Preparation of the fortified tomato purees on lab scale

For fortified tomato purees preparation, the GS-L, GS-M and GS-S fractions (3.2 g) were added to the PV and PR tomato purees (96.8 g) to achieve 3% fibre content in the final product. Each puree was filled into different glass bottles (250 mL capacity). A set of the bottled fortified purees and their corresponding control purees were then submitted to microwave heating (8 min at 900 watt). During heating, the temperature of the tomato puree was monitored by using a thermocouple set in the geometric centre of one of the bottles (the slowest heating point) to attain the heating curve and calculate the pasteurisation effect (Earle and Earle 2003). Six decimal reductions (6D) of the target microorganism *A. acidoterrestris* (D_{ref} = 1.5 min, T_{ref} =95 °C and z-value = 7 °C, Bevilacqua and Corvo, 2011) was achieved, which can be considered effective for acidic foods (Silva and Gibbs 2004). In parallel, another set of the bottled fortified purees and the PV and PR controls were submitted to autoclave treatment (100 °C, 30 min). Discontinuous and intensive autoclave treatments are also common in tomato processing industry (Figure 1).

Moisture and Dietary Fibre

Moisture content of tomato purees and GS was determined by drying samples in a vacuum oven at 70 °C and 50 Torr for 18 h (AOAC 1990). Dietary fibre content was determined by using the procedure described in the Megazyme (Megazyme International Ltd, Bray, Ireland) total dietary fibre assay kit, which is based on AOAC 991.43 (AOAC 1990).

Sample Extraction

For GS extraction, 1 g was weighted, added with 20 mL methanol:water:formic acid (70:29.9:0.1, v/v/v) and extracted for 2 h at 60 °C with continuous stirring. The mixture was centrifuged at 10000g for 10 min, the supernatant recovered and the solid residue was re-extracted using 10 mL of the same solvent. The supernatants were pooled. For tomato puree extraction, 3.75 g was weighted and added with 1.9 mL of water, 7 mL of methanol and 0.3 mL of formic acid (in order to use the same medium as for the GS fractions, taking into account the amount of water present in the puree). Extractions were performed as that of the GS fractions. Extracts were stored at -20°C until analytical characterisation.

Total soluble phenolics

The Folin–Ciocalteu assay was performed as described previously (Singleton et al., 1999). The reaction mixture contained 6.0 mL of distilled water, 0.5 mL of the extracts diluted with methanol:water:formic acid (70:29.9:0.1, v/v/v), 0.5 mL of Folin–Ciocalteu reagent and 3 mL of 10% Na_2CO_3. The mixtures were incubated for 90 min at room temperature and then the absorbance was recorded at 760 nm against a blank with no extract addition using a UVdec-610 spectrophotometer (Jasco, Lecco, Italy). For each extract, 2 - 4 dilutions were assessed. A calibration curve was built using gallic acid. Total soluble phenolics were expressed as milligrams of gallic acid equivalents (GAE) per kilogram of product.

Particle size distribution (PSD)

The analysis of PSD of tomato purees in the range 0.2-1000 μm, GS granulometric fractions and their mixtures was performed according to the specifications reported in the international standard ISO 13320 (2009) using a Malvern 2000 Laser granulometer (Malvern instruments Ldt., Malvern, UK) equipped with a single laser source at $\lambda = 633$ nm. Deionised water was used as the dilution medium (1:500 ratio) in order to avoid multiple scattering phenomena and excessive obscuration of the laser source; the temperature was set at 25°C ± 1°C. The high dilution factor used and sonication of diluted sample allowed to neglect the contribution of particle coalescence. The refractive index for water used as dispersant medium was set to 1.33, and in order to model the angular intensity distribution function of scattered light, the Fraunhofer approximation was adopted for all samples, described as:

$$\frac{I(\theta)}{I_0} = \frac{1}{k^2 l_a^2}\alpha^4\left[\frac{J_1 \alpha \sin\theta}{\alpha \sin\theta}\right]^2 \qquad (1)$$

where: $I(\theta)$ is the angular intensity distribution of light scattered by particles, I_0 is the intensity of the incident unpolarised light, k is the wavenumber in the medium, l_a is the distance from scattering object to detector, α is the dimensionless size parameter ($\alpha = \pi x n_m / \lambda$, with x, particle size, n_m, refractive index of the medium, λ wavelength of illuminating light source in vacuum),

J_1 is first order Bessel function, ϑ is the scattering angle with respect to forward direction. Fraunhofer approximation is valid for these samples because their particle sizes were much bigger than λ (ISO 13320, 2009). Calculations for PSD and its descriptors were made using the instrument's software General Purpose Model, assuming a spherical particle shape with normal calculation sensitivity. The descriptors considered were the surface-weighted mean diameter (μm), i.e., d(3,2), also called Sauter mean diameter, the volume moment-weighted mean diameter (μm) or d(4,3), defined as:

$$\mathrm{d}(3,2) = \frac{\sum_i n_i d_i^3}{\sum_i n_i d_i^2} \qquad (2)$$

$$\mathrm{d}(4,3) = \frac{\sum_i n_i d_i^4}{\sum_i n_i d_i^3} \qquad (3)$$

where d_i is the i-th diameter class and n_i is the respective number of particles per unit volume, and the width of the distribution, i.e., span, defined as:

$$\mathrm{Span} = \frac{(\mathrm{d}0.9 - \mathrm{d}0.1)}{\mathrm{d}0.5} \qquad (4)$$

where d0.1, d0.9 and d0.5 are 10, 50 and 90% quantile, respectively.

The PSD obtained using a laser diffraction method is expressed in terms of equivalent spheres on volume basis.

Bostwick consistency

Consistency was determined with a Bostwick consistometer (LS 100, Labo-Scientifica, Parma, Italy). Measurements taken after both 30 s and 60 s gave the same information. Results were expressed as distance travelled by the sample (cm) through the trough in 60 s.

Rheological properties

The fundamental rheological properties of raw and fortified tomato purees were studied by means of dynamic oscillatory measurements performed on a Physica MCR300 Rheometer (Anton Paar GmbH, Graz, Austria), supported by the software Rheoplus/32 (v. 3.00, Physica Messtechnik GmbH, Ostfildern, Germany). A parallel plate geometry (25 mm diameter, 2 mm gap) was used, with corrugated plates to prevent sample slippage. The temperature was regulated at 25 °C by using a circulating bath and a controlled Peltier system. After loading, the excess sample was trimmed off, and before starting the tests, the sample was allowed to rest for 5 min to relax stress. A moisturizing external chamber was used to prevent moisture loss during measurements.

The fundamental rheological properties were determined within the linear viscoelastic region (0.01-0.2% strain), as determined by the amplitude sweep performed in the range of 0.01-300% strain, at a constant frequency of 1

Hz. Frequency sweep was carried out under a constant strain of 0.1% in the range of frequencies 0.1-10 Hz to calculate the elastic modulus (G' , Pa), the viscous modulus (G'', Pa) and complex viscosity (η*, Pa·s), defined as:

$$\eta * = \sqrt{\frac{(G\prime)^2 + (G\prime\prime)^2}{\varpi}} \qquad (5)$$

where ϖ is the angular oscillatory frequency (rad/s) .

Colorimetric parameters

Colour evaluation was performed in quadruplicate with a Chroma meter II (Konica Minolta, Osaka, Japan), which provides the Hunter $L*$, $a*$, and $b*$ coordinates, representing: lightness and darkness ($L*$), redness ($+a*$), greenness ($-a*$), yellowness ($+b*$), and blueness ($–b*$). The calibration of chromameter was checked with a standard white reflective plate. The head of the colorimeter was placed directly on the bottom of the bottles containing the tomato purees for colour measurement.

To study the total variation in colour, colour difference, namely ΔE_T were calculated, as indicated by the following equation:

$$\Delta E_T = \sqrt{(a^* - a^*_{UH})^2 + (b^* - b^*_{UH})^2 + (L^* - L^*_{UH})^2} \qquad (6)$$

where $a*$, $b*$, and $L*$ are the values of the colorimetric parameters of heat-treated purees and $a*_{UH}$, $b*_{UH}$, and $L*_{UH}$ are the values of the colorimetric parameters of the corresponding unheated tomato puree.

Statistical Analysis of Data

Experimental data were obtained in triplicate and analysed by one-way ANOVA using the least significant difference (LSD) as a multiple range test, and by linear regression analyses using Statgraphics 5.1 (STCC Inc.; Rockville, MD, USA). Results are reported as average ± SD.

Results and Discussion

Particle size distribution (PSD)

Tomato puree generally forms structured suspensions consisting of cells (mainly parenchyma cells) and/or cell wall material dispersed and arranged in a liquid matrix phase, which is comprised of soluble materials such as polysaccharides, i.e., pectins and sugars, and some proteins (Bayod & Tornberg, 2011).

In the current study, PV and PR tomato purees were fortified with antioxidant- and fiber-rich GS and six formulations having various particle sizes were obtained. PSD was then studied in the unfortified purees and their formulations with GS. PSD of control purees showed polydisperse monomodal behaviours (Figure 2) with different d(3,2) values (122.8 ± 0.4 and 185.7 ± 0.6 µm for PV and PR, respectively) and d(4,3) values (323.5 ±1.0 and 392.0 ± 2.3 µm for PV and PR, respectively). PV showed a wider span (1.87 ± 0.01) than PR (1.74 ± 0.01) due to smaller particle fractions. Hence, the PV and PR samples chosen for the experimental set-up can be considered as different models of puree. Both d(3,2) and d(4,3) values did not correspond to the sieves used during wet sieving of purees (0.5 mm for PV and 1 mm for PR, as shown in Fig. 1). Accordingly, in a previous study, the d(4,3) values of tomato pastes have been reported to be in the range between 324 and 467 μm and it was found that they do not correspond to the sieves used during processing (0.8 – 2.2 mm) (Sanchez et al., 2002). In addition, the finisher did not remove all the particles with maximum diameter greater than 1 mm. In fact, PSD up to 2 mm were found, however their presence was very low (Figure 2). This result was also observed previously and was explained by the fact that parenchyma cells, accounting for the majority of cells in vegetable suspensions, are highly deformable. In addition, tomato-derived particles are non-spherical. Hence, they can pass through pores with a smaller size than the size of the particle itself (Moelants et al., 2014a). As clarified in the following paragraph, these parameters were also affected by heat-treatment.

With reference to GS fractions, GS-L and GS-M showed monomodal PSD (Figure 2) with mean d(3,2) of 66.0 ± 0.8 and 33.1 ± 0.2 μm, respectively, while GS-S showed bimodal PSD with mean d(3,2) of 18.0 ± 0.1μm due to the presence of finer particle size. However, GS-L and GS-M also contained a fraction of fine particles, greatly influencing the d(3,2) values. Median diameters on volumetric basis (d0.5) of the three GS fractions were very similar to the values of d(4,3) (not shown) and equal to 516.1 ±7.5, 235.9 ± 1.2 and 64.1 ± 0.2 μm for GS-L, GS-M and GS-S respectively, which were consistent with the sieves' meshes adopted during dry sieving.

As shown in Table 1, after the mixing step, it was possible to notice that d(3,2) appeared to be very much influenced by the addition of the GS-S fraction, while the variations induced by GS-L and GS-M were minor and not significant for PVGS-L puree. For the PV purees, control PV, PVGS-L, PVGS-M and PVGS-S purees had d(3,2) values of 122.8 ± 0.4, 116.0 ± 3.8, 110.6 ± 4.3 and 93.0 ± 2.9 μm, respectively. A similar trend was observed for the mean d(4,3) diameter, which highlighted considerable changes for PVGS-S puree. PV, PVGS-L, PVGS-M and PVGS-S purees had d(4,3) of 323.5 ± 1, 335.7 ± 4.6, 317.4 ± 4.1 and 311.0 ± 3.2 μm, respectively. This parameter, which is mainly influenced by the larger particles of the formulation, showed therefore smaller variations than the Sauter mean diameter. The span of the PSD was significantly modified only by the addition of GS-S in both PV and PR purees.

The effect of heat treatment on the PSD was observed as a decrease of the Sauter mean diameter for all PR purees and PVGS-S puree, but not for PVGS-M and PVGS-L purees. Both tomato puree and grape skins particles are made of plant cell wall material. The particles in plant-tissue based food suspensions are built up of mechanically destructed parenchyma tissue. Parenchyma cells are glued together by the pectin of the middle lamella. Thermal treatments cause a weakening of such tissues by beta-eliminative pectin degradation or acid hydrolysis of pectin (Moelants et al., 2014b). This could explain the observed decrease in d(3,2), i.e., particle fragmentation. The d(4,3) parameter, which is mostly affected by the large particles, increased significantly for the purees added with GS-M and GS-L fractions, but did not show significant changes for those added with the GS-S fraction. When sheared tomato suspensions are kept in quiescent conditions for a certain time, additional connections can be formed between the particles (Moelants et al., 2014a). Hence, it can be hypothesized that in quiescent conditions after heat treatments caused further aggregation, as observed by increased d(4,3). The span turned out to be less affected by heat treatments.

Dietary Fibre and Total Soluble Phenolics

The incorporation of different grape skin varieties into a tomato puree can bring about considerable changes in the chemical properties due to variability with respect to phenolic composition and, to a lesser extent, in fibre content (Quen et al., 2011). In the current study, Chardonnay was chosen as a model variety since it is one of the oldest and most widely distributed wine grape cultivars and is of commercial importance for the world‘s wine-producing nations (Gambetta et al., 2014). Fibre content of Chardonnay skin was found to be 50.5%. In previous food fortification studies with GS, the dietary content of GS ingredient was ~ 50% (Mildner-Szkudlarz et al., 2011; Mildner-Szkudlarz et al., 2013; Riberio et al., 2013; Tseng and Zhao, 2013), since this level is necessary to define it as an “antioxidant dietary fibre” (Saura-Calixto, 1998). Dietary fibre content of PV and PR purees was 1.5 ± 0.1%. The level of addition of GS to PV and PR purees was chosen in order to increase dietary fibre content to 3.0 ± 0.1%. Hence, the formulated purees can be labelled as “fibre-source” according to the EU Regulation N. 1924/2006.

The amount of total soluble phenolic compounds were 24 ± 1, 22 ± 1 and 18 ± 1 g/kg in the GS-S, GS-M and GS-L fractions, respectively. The higher level found in the GS-S fraction, with respect to GS-M and GS-L, was probably attributable to a better extraction yield due to increased surface/solvent contact area in the GS-S fraction that had smaller particle sizes than the GS-M and GS-L fractions. In fact, besides low molecular weight soluble phenolics, GS contain a large amount of high molecular weight proanthocyanidins, which have low solubility and can be quantified only after acidic depolymerisation (Sri Harsha et al. 2013).

Total soluble phenolics of the PR and PV purees (6.52 – 7.13 g GAE/kg) (Table 2) were in the range of those observed for twenty cultivars of fresh tomatoes extracted with an optimised procedure (Li et al. 2011). After addition of GS fractions to tomato purees, the total soluble phenolic content increased, leading to potential health benefits of the innovative purees with respect to those of the conventional purees. Chardonnay variety, which is used worldwide, was found to have low phenolic content as compared to other white grape skins (Sri Harsha et al., 2014). Hence, an increase in phenolic content in the fortified purees can also be expected upon addition of skins of other grape varieties.

In general, total soluble phenolic content in the fortified purees were not affected by the microwave treatment, but increased after autoclave treatment, which also caused decrease in d(3,2), i.e., increased surface/solvent contact. Indeed in the fortified purees, total soluble phenolic content were inversely correlated to the d(3,2) values ($R = -0.79$, p 0.0001, Figure 3). It is worth noting that, one of the advantages of using GS as ingredient is linked to the delivery of grape-phenolics. Hence, as a general rule, the increase in grape phenolic solubility with decreasing GS particle size and with intense thermal treatment should be taken into account to enhance the nutritional and functional properties of grape phenolics. In particular, grape phenolics have proven health benefits (Perez-Jimenez, 2008; Lopez-Oliva et al., 2010). In addition, grape phenolics were found to prevent oxidation in fish (Riberio et al., 2013) and meat (Sáyago-Ayerdi et al. 2009) and to act as antifungal agents in apple and orange juices (Sagdic et al., 2011). Hence, these compounds could replace synthetic additives.

Bostwick consistency

The flow properties of tomato products referred to as the gross viscosity or the consistency, are typically evaluated using a Bostwick consistometer. It should be noted that more viscous juices flow shorter distances and thus, higher Bostwick value indicates a lower consistency. Bostwick value is a fundamental quality parameter for tomato derivatives and loss of consistency has to be considered as a negative effect (Anthon et al. 2010).

PV and PR, which were concentrated and stored in tank before heat treatment had low consistency values of 8.7 and 7.2 cm, respectively (Table 3). Indeed, for a puree which is processed on the same day of harvest and not submitted to concentration step, consistency value was found to be around 6 cm (not shown). During tomato concentration there is a loss of consistency, which is maximum for tomato paste. One proposed mechanism for consistency loss in tomato paste is that the high osmotic and ionic strength in the paste causes changes in the polymeric materials in juice particles, altering the interactions between these particles (Anthon et al. 2010).

Upon mixing tomato purees with GS fractions (3.2 g), the total dry solids increased and a statistically significant rise in consistency was observed in PV and PR purees (Table 3). Interestingly in the fortified purees, consistency values were in the range 4.7 – 6.0 cm. This result deserves practical interest as a similar range for Bostwick consistency values (2.5 – 5.5 cm) has been obtained for ketchup samples added with chemically modified starches as thickening agents, which increase the viscosity of serum (Juszczak et al., 2013).

In the formulated purees having the same tomato matrix (either PV or PR), the particle sizes played a major role in consistency changes. After mixing, purees added with GS-S fraction exhibited higher increase than GS-L formulated purees, while GS-M formulated purees exhibited an intermediate behaviour (Table 3). A larger surface might enable additional interactions with the water phase, thus leading to increased viscosity of the serum as observed when modified starches are added to tomato paste (Juszczak et al., 2013). GS particles can fill the space between tomato fragments and trap more water, indeed dietary fibre possess water holding and gel-forming ability (Elleuch et al., 2011). In addition, a larger surface might enable more physical entanglements among polymers (Moelants et al. 2014a).

Upon both the microwave and the autoclave treatments, a further statistically significant increase in consistency was observed in most of the fortified purees and their corresponding controls, especially in GS-S-formulated purees. In a previous study, it was hypothesized that modifications that contribute to increased component solubilisation, such as decreased particle size and a rise in temperature, cause an increase in viscosity (Cordoba et al. 2012). Accordingly in the present study, decrease in d(3,2) values suggests that disaggregation of the particles occurred upon heat treatments, thus leading to increased interaction between solids and the water phase and further entanglements of polymers as observed for concurrent increase of d(4,3) values.

Rheological properties

The oscillatory frequency sweep tests were performed at a constant strain of 0.1%, which was found to be within the linear viscoelastic region for all the control and fortified purees, in the frequencies 0.1 - 10 Hz (Figure 4).

The critical parameters that influence the rheological properties of tomato concentrate are the tomato variety, the sieve pore sizes, processing temperature and the volume fraction of solids (Juszczak et al. 2013), while the serum viscosity is of secondary importance (Moelants et al. 2013). All tomato purees showed low viscoelastic behaviour, as G' (storage modulus) dependency of oscillatory frequency was very small. Values of G' were always higher than those of G'' (loss modulus), which indicates that tomato purees had dominant elastic properties rather than viscous behaviour. Thus, the products can be classified as weak gels (Rao 1999). This behaviour is typically observed in fruit products, as reported for peach puree (Massa et al. 2010), peach juice with fibres (Augusto et al., 2011), and tomato concentrates (Bayod & Tornberg, 2011).

Control PV and PR tomato purees showed the same general trend as described, but different magnitudes for both G’ and G’’ values, in the following order: PR > PV (Figure 4). Similar sequence was observed for Bostwick consistency.

After addition of the GS fractions, similar mechanical spectra were observed with respect to the controls, but G’ and G’’ were both increased sharply, most likely related to increased dry solids in the puree. The same effect was observed by Tiziani et al. (2005) upon addition of soy protein to tomato juice. The reason was explained as enhanced non-covalent aggregation among the macromolecules that increased the stability of the suspension. The values of G’, calculated at 1 Hz, increased from 407 ± 57 Pa in control PV to ~ 935 ± 47 Pa in the GS-formulated PV purees. Similarly, an increase in G’ values was noticed from 788 ± 240 Pa in control PR puree to ~ 1763 ± 105 Pa in GS-formulated PR purees (Figure 4).

The values of G”, calculated at 1 Hz increased from 61 ± 10 Pa in the control PV puree to ~151 ± 21 Pa in the GS-formulated PV purees and from 136 ± 51 Pa in control PR puree to ~ 346 ± 24 Pa in the GS-formulated PR purees (Figure 4). Heat treatments did not affect G’ and G’’ significantly (not shown).

The complex viscosity (η*) is the frequency-dependent viscosity function determined during forced harmonic oscillation of shear stress. Values for η*, were then calculated at 1 Hz, to further investigate the effect of GS addition to tomato puree (Figure 5). These values increased from 60 ± 9 Pa.s in the control PV puree to ˗134 ± 11 Pa.s in the GS formulated purees and from 118 ± 36 Pa.s in the control PR puree to ˗ 248 ± 42 Pa.s in the GS formulated purees. Hence, significant increases in η* values were observed when the control purees were added with various GS fractions. No significant changes occurred upon heating, except for PVGS-S.

In the fortified purees, η* values appeared to be correlated with d(4,3) values (R = 0.86, P = 0.0000) (Fig. 2), supporting the occurrence of enhanced non-covalent aggregation among the fibres added and tomato particles as already suggested (Tiziani et al. 2005).

Some previous applications of GS as ingredients have related the observed changes of mechanical properties to the granulometry of this novel ingredient. GS with particle sizes of 0.18 mm was selected for the fortification of yogurt and salad dressing, which caused a positive increase in viscosity (Tseng and Zhao, 2013). On the contrary, GS decreased the instrumental and sensory perceived elasticity of fish, thus leading to unpleasant texture, and this effect was ascribed to the large granulometry, i.e., particle sizes ≤ 1 mm (Riberio et al., 2013). For meat products, GS having particle sizes ≤ 0.5 mm were selected, which did not cause changes in tenderness (Sáyago-Ayerdi et al. 2009). Other studies have evidenced positive effect of using GS as an ingredient to improve gel properties in milk for cheese manufacturing (Han et al., 2011) and to enhance the hardness and gumminess of a rye bread (Mildner-Szkudlarz et al., 2011), however the granulometry of the ingredient was not specified.

Colorimetric parameters

Colour is one of the most important quality characteristics for the tomato processing industry, since it has an important influence on consumers' preference. Changes in colour due to sterilisation of tomato puree are due to non-enzymatic browning, whereas the typical red carotenoid lycopene is heat-stable (Zanoni et al. 2003). Grape fibre addition is also expected to modify food colour, as previously observed for GS added to chicken breast hamburger (Sayago-Ayerdi et al. 2009) and to an ice cream formulation (Sagdic et al., 2011).

Indeed, relevant variations in L^* and a^* values were observed in tomato purees upon mixing with GS fractions (Table 4), whereas changes of b^* values were less (not shown). Variation in L^* and a^* parameters were higher after addition of the GS-S fractions than that of GS-M and GS-L fractions. Higher L^* values in the purees added with GS-S fractions are probably due to backscattering of particles with diameter less than a 30 µm, causing white colour to their suspensions (Walstra 2003).

After the heat treatments, a^* values increased for all the fortified samples, and moderately decreased for the controls. L^* values increased in the fortified tomato purees; this result could be an effect of increase of small particles, as observed from decrease in d(3,2).

The total colour difference was also calculated using the sample before heat treatment as a reference (ΔE_T), as reported previously. Values between 1.0 and 3.1 were observed for the microwaved purees, whereas values between 2.0 and 3.0 were observed for the autoclaved purees. Accordingly, ΔE_T values in the range 2.42 - 3.79 were found by Giner et al. (2013) upon tomato juice pasteurisation. These values indicate that the extent of colour modification in the fortified purees was low. In fact, in a previous study, a ΔE_T value of 5 was considered as a threshold for acceptable colour variation during tomato sterilisation (Zanoni et al. 2003).

Conclusions

Use of GS as a healthy ingredient for low energy dense tomato purees can lead to various formulations with different physical properties, depending on the particle sizes. As a general rule, addition of the antioxidant- and fibre-rich GS ingredient increased consistency and gel properties of tomato purees.

The d(3,2) of the GS ingredient was inversely correlated to phenolic solubility, which is associated to the potential nutritional and functional properties (antioxidant, antifungal agents) of the new formulations. Bostwick consistency, G’, G’’ and η* increased in the fortified purees. The η* values displayed a positive correlation with d(4,3) values. As shown by the above-mentioned correlations, the addition of specific granulometric fractions of GS could be used for tuning nutritional, functional and physical properties in tomato purees. The results obtained from this methodological approach could pave way for the optimisation of fibrous by-products incorporation into complex food fluids.

List of symbols

Symbol	Caption	Unit
d(3,2)	surface-weighted mean diameter (Sauter mean diameter)	µm
d(4,3)	volume-weighted mean diameter	µm
d0.1, d0.5 and d0.9	10%, 50% and 90% quantile of particle size distribution on a volumetric basis	µm
G’	storage modulus	Pa
G’’	loss modulus	Pa
η^*	complex viscosity	Pa·s
Ω	angular oscillatory frequency	rad/s
D	decimal reduction time	min
Z	temperature interval causing a 10-times variation in the D-value	°C
ϑ	scattering angle with respect to forward direction	°
$I(\vartheta)/I_0$	Ratio of angular intensity distribution of light scattered by particles and intensity of the incident unpolarised light	
K	wavenumber in the medium	nm^{-1}
l_a	the distance from scattering object to detector	mm
A	dimensionless size parameter	
X	particle size	µm
λ	wavelength of illuminating light source in vacuum	nm
J_1	first order Bessel function	
n_m	refractive index of the medium (water)	
L^*	Hunter’s lightness index	
a^*	Hunter’s redness index	
b^*	Hunter’s blueness index	
ΔE_T	Total variation in Hunter’s colorimetric parameters	

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Figure captions

Fig. 1. Schematic Overview of Tomato Processing to Fortified Purees. Top section: Industrial Processing of PV and PR Purees. Bottom section: Lab Scale Mixing of PV and PR Purees with GS Fractions Followed by either

Optimized Heat Stabilisation (Microwave Treatment) or Intensive Heating (Autoclave Treatment). Asterisks (*) Indicate the Sampling Points. Abbreviations: PV, Smooth Puree (≤ 0.5 mm); PR, Rough Puree (≤ 1 mm); GS, Granulometric Fractions of Dried Grape Skins: GS-L (250µm < GS-L ≤ 500µm), GS-M (125µm < GS-M ≤ 250µm) and GS-S (GS-S ≤ 125µm).

Fig. 2. PSD of PV and PR Tomato Purees and Grape Skins Granulometric Fractions GS-S; GS-M and GS-L. Abbreviations are the Same as in Fig. 1.

Fig 3. Correlations between Surface-Weighted Mean Diameter, d(3,2) and Soluble Phenolics (g GAE/kg) and between Volume-Weighted Mean Diameter, d(4,3) and Complex Viscosity for all the Fortified Purees Subjected to Different Treatments (Mixing, Microwave, Autoclave).

Fig. 4. Frequency Sweep Test: Variation in the Storage Modulus (G', Pa) and Loss Modulus (G'', Pa) of Tomato Purees Formulated with GS Fractions and Control Tomato Purees. Abbreviations are the Same as in Fig.1. (n = 3).

Fig. 5. Complex Viscosity (η*, Pa·s) values (at 1 Hz) of Tomato Purees Formulated with GS Fractions and Control Tomato Purees after Mixing and Autoclave Treatments. Different letters within the same treatment (a –e) or puree (x – y) indicate significant differences (LSD, $p < 0.01$). Abbreviations are the Same as in Fig.1. Error bars indicate SD. (n = 3).

Table 1. Surface-Weighted Mean Diameter (d(3,2), μm), Volume -Weighted Mean Diameter (d(4,3), μm) and Span of Tomato Purees Formulated with GS Fractions and Control Tomato Purees after Mixing, Microwave and Autoclave Treatments.

Puree	**d(3,2)**			**d(4,3)**			**Span**		
	Mixing	**Microwave**	**Autoclave**	**Mixing**	**Microwave**	**Autoclave**	**Mixing**	**Microwave**	**Autoclave**
PV (control)	$122.8^{xcd} \pm 0.4$	$122.1^{xc} \pm 1.8$	$123.6^{xe} \pm 0.5$	$323.5^{xb} \pm 1.0$	$327.7^{xb} \pm 5.6$	$331.1^{xc} \pm 1.1$	$1.87^{xb} \pm 0.01$	$1.86^{xb} \pm 0.02$	$1.89^{xb} \pm 0.02$
PVGS-L	$116.0^{xbc} \pm 3.8$	$115.2^{xbc} \pm 0.8$	$113.5^{xd} \pm 0.1$	$335.7^{xc} \pm 4.6$	$349.5^{yc} \pm 2.2$	$348.3^{yd} \pm 0.9$	$1.90^{xbc} \pm 0.02$	$1.94^{xyc} \pm 0.01$	$1.95^{yc} \pm 0.01$
PVGS-M	$110.6^{xb} \pm 4.3$	$109.7^{xb} \pm 14.5$	$99.9^{xb} \pm 0.6$	$317.4^{xab} \pm 4.1$	$326.6^{ybx} \pm 6.7$	$323.4^{ybx} \pm 1.6$	$1.87^{xb} \pm 0.02$	$1.89^{xbc} \pm 0.02$	$1.87^{xb} \pm 0.02$
PVGS-S	$93.0^{ya} \pm 2.9$	$82.0^{xa} \pm 0.4$	$81.3^{xa} \pm 0.6$	$311.0^{xa} \pm 3.2$	$312.0^{xa} \pm 1.6$	$314.2^{xa} \pm 3.5$	$2.02^{xd} \pm 0.02$	$2.04^{xd} \pm 0.01$	$2.09^{yd} \pm 0.02$
PR (control)	$185.7^{yg} \pm 0.6$	$177.5^{xf} \pm 4.7$	$170.9^{xh} \pm 1.1$	$392.0^{xe} \pm 2.3$	$400.8^{yf} \pm 3.4$	$396.5^{xyg} \pm 1.0$	$1.74^{xa} \pm 0.01$	$1.73^{xa} \pm 0.01$	$1.75^{xa} \pm 0.01$
PRGS-L	$173.2^{zf} \pm 1.7$	$158.4^{ye} \pm 0.7$	$152.1^{xg} \pm 2.5$	$393.6^{xe} \pm 3.0$	$416.0^{zg} \pm 1.6$	$404.7^{yh} \pm 3.7$	$1.74^{xa} \pm 0.01$	$1.72^{xa} \pm 0.01$	$1.74^{xa} \pm 0.01$
PRGS-M	$150.7^{ye} \pm 1.2$	$145.3^{yd} \pm 5.0$	$133.5^{xf} \pm 1.5$	$374.4^{xd} \pm 3.5$	$387.3^{ye} \pm 7.1$	$386.9^{yf} \pm 4.6$	$1.77^{xa} \pm 0.01$	$1.72^{xa} \pm 0.03$	$1.75^{xa} \pm 0.01$
PRGS-S	$124.2^{zd} \pm 1.2$	$111.7^{ybc} \pm 1.2$	$105.7^{xc} \pm 0.2$	$366.6^{xd} \pm 3.0$	$374.7^{yd} \pm 2.4$	$368.8^{xe} \pm 1.9$	$1.94^{yc} \pm 0.02$	$1.91^{xc} \pm 0.01$	$1.98^{zc} \pm 0.01$

Results are the average values ± SD (*n* = 3). Different letters within the same column (a – h) or row (x – z) indicate significant differences (LSD, $p < 0.05$). Abbreviations are the same as in Fig.1.

Table 2. Total Soluble Phenolics (gGAE/kg) of Tomato Purees Formulated with GS Fractions and Control Tomato Purees after Mixing, Microwave and Autoclave Treatments.

Puree	**Total Soluble Phenolics**		
	Mixing	**Microwave**	**Autoclave**
PV (control)	7.13^{xa} ± 0.03	7.49^{xab} ± 0.72	7.72^{xb} ± 0.09
PVGS-L	8.85^{xc} ± 0.42	9.75^{xcde} ± 0.99	10.17^{xc} ± 0.01
PVGS-M	9.80^{xd} ± 0.18	10.17^{xde} ± 0.21	11.05^{yd} ± 0.18
PVGS-S	10.28^{xd} ± 0.41	10.89^{ye} ± 0.01	11.00^{yd} ± 0.11
PR (control)	6.52^{xa} ± 0.16	6.37^{xa} ± 0.02	6.16^{xa} ± 0.26
PRGS-L	8.15^{xb} ± 0.38	8.55^{xybc} ± 0.22	9.68^{yc} ± 0.56
PRGS-M	8.78^{xbc} ± 0.30	9.29^{xycd} ± 0.44	10.19^{yc} ± 0.01
PRGS-S	9.73^{xd} ± 0.34	10.03^{xde} ± 0.54	11.90^{ye} ± 0.54

Results are the average values ± SD ($n = 3$). Different letters within the same column (a – e) or row (x, y) indicate significant differences (LSD, $p < 0.05$). Abbreviations are the same as in Fig.1.

Table 3. Bostwick Consistency (cm in 60 s) of Tomato Purees Formulated with GS Fractions and Control Tomato Purees after Mixing, Microwave and Autoclave Treatments.

Puree	**Bostwick consistency**		
	Mixing	**Microwave**	**Autoclave**
PV (control)	$8.7^{yf} \pm 0.4$	$7.4^{xe} \pm 0.5$	$7.8^{xf} \pm 0.4$
PVGS-L	$6.0^{yd} \pm 0.4$	$5.4^{xyc} \pm 0.2$	$4.8^{xd} \pm 0.2$
PVGS-M	$5.9^{zcd} \pm 0.1$	$4.8^{yabc} \pm 0.0$	$4.2^{xc} \pm 0.1$
PVGS-S	$5.3^{zbc} \pm 0.1$	$4.6^{yabc} \pm 0.2$	$3.7^{xb} \pm 0.1$
PR (control)	$7.2^{ye} \pm 0.1$	$6.3^{xd} \pm 0.7$	$6.8^{xye} \pm 0.3$
PRGS-L	$5.3^{xab} \pm 0.3$	$4.9^{xbc} \pm 0.2$	$5.0^{xd} \pm 0.3$
PRGS-M	$4.7^{ya} \pm 0.1$	$4.3^{xab} \pm 0.0$	$4.2^{xc} \pm 0.1$
PRGS-S	$4.7^{ya} \pm 0.4$	$4.0^{xa} \pm 0.0$	$3.3^{xa} \pm 0.1$

Results are the average values ± SD (*n* = 3). Different letters within the same column (a – f) or row (x – z) indicate significant differences (LSD, $p < 0.05$). Abbreviations are the same as in Fig.1.

Table 4. Colorimetric parameters *L** and *a** and ΔE_T of Tomato Purees Formulated with GS Fractions and Control Tomato Purees after Mixing, Microwave and Autoclave Treatments.

Puree	***L****			***a****			**ΔE_T**	
	Mixing	**Microwave**	**Autoclave**	**Mixing**	**Microwave**	**Autoclave**	**Microwave**	**Autoclave**
PV (control)	42.4^{yb} ± 0.1	42.4^{yab} ± 0.1	41.3^{xa} ± 0.1	16.8^{ye} ± 0.1	15.0^{xd} ± 0.1	16.5^{yd} ± 0.1	3.1^{c}	2.0^{a}
PVGS-L	41.4^{xa} ± 0.1	41.7^{xa} ± 0.1	43.8^{yc} ± 0.1	12.3^{xb} ± 0.1	14.0^{yc} ± 0.1	14.0^{yabc} ± 0.1	1.7^{abc}	2.9^{ab}
PVGS-M	42.2^{xb} ± 0.1	43.2^{ybc} ± 0.1	44.1^{zc} ± 0.1	12.5^{xb} ± 0.1	13.0^{yb} ± 0.1	14.5^{zc} ± 0.1	1.7^{abc}	3.0^{b}
PVGS-S	45.0^{xd} ± 0.1	44.4^{yc} ± 0.1	45.8^{ze} ± 0.1	12.9^{xc} ± 0.1	13.0^{xb} ± 0.1	14.3^{ybc} ± 0.1	1.0^{a}	2.2^{ab}
PR (control)	43.4^{xc} ± 0.1	44.6^{yc} ± 0.1	42.5^{xb} ± 0.1	16.3^{yed} ± 0.1	14.9^{xd} ± 0.1	15.8^{xyd} ± 0.1	2.2^{c}	2.0^{a}
PRGS-L	42.2^{xb} ± 0.1	43.7^{ybc} ± 0.1	44.3^{yc} ± 0.1	11.8^{xa} ± 0.1	12.4^{xya} ± 0.1	13.3^{ya} ± 0.1	1.6^{ab}	2.8^{ab}
PRGS-M	43.4^{xc} ± 0.1	44.3^{yc} ± 0.1	45.0^{yd} ± 0.1	11.8^{xa} ± 0.1	12.2^{xa} ± 0.1	13.4^{yab} ± 0.1	1.0^{a}	2.4^{ab}
PRGS-S	45.9^{xe} ± 0.1	46.9^{yd} ± 0.1	47.3^{yf} ± 0.1	12.6^{xbc} ± 0.1	12.9^{xb} ± 0.1	13.9^{yabc} ± 0.1	1.0^{a}	2.5^{ab}

Results are the average values ± SD (*n* = 3). Different letters within the same column (a – f) or row (x – z) indicate significant differences (LSD, $p<0.05$). ΔE_T is the difference in consistency between unheated and heated puree. Abbreviations are the same as in Fig.1.

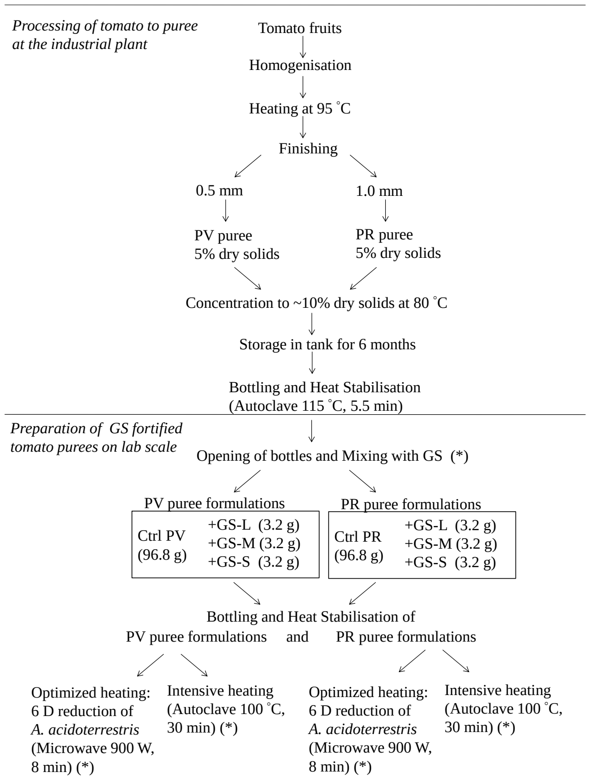

Fig. 1. Schematic Overview of Tomato Processing to Fortified Purees. Top section: Industrial Processing of PV and PR Purees. Bottom section: Lab Scale Mixing of PV and PR Purees with GS Fractions Followed by either Optimized Heat Stabilisation (Microwave Treatment) or Intensive Heating (Autoclave Treatment). Asterisks (*) Indicate the Sampling Points. Abbreviations: PV, Smooth Puree (≤ 0.5 mm); PR, Rough Puree (≤ 1 mm); GS, Granulometric Fractions of Dried Grape Skins: GS-L (250µm < GS-L ≤ 500µm), GS-M (125µm < GS-M ≤ 250µm) and GS-S (GS-S ≤ 125µm).

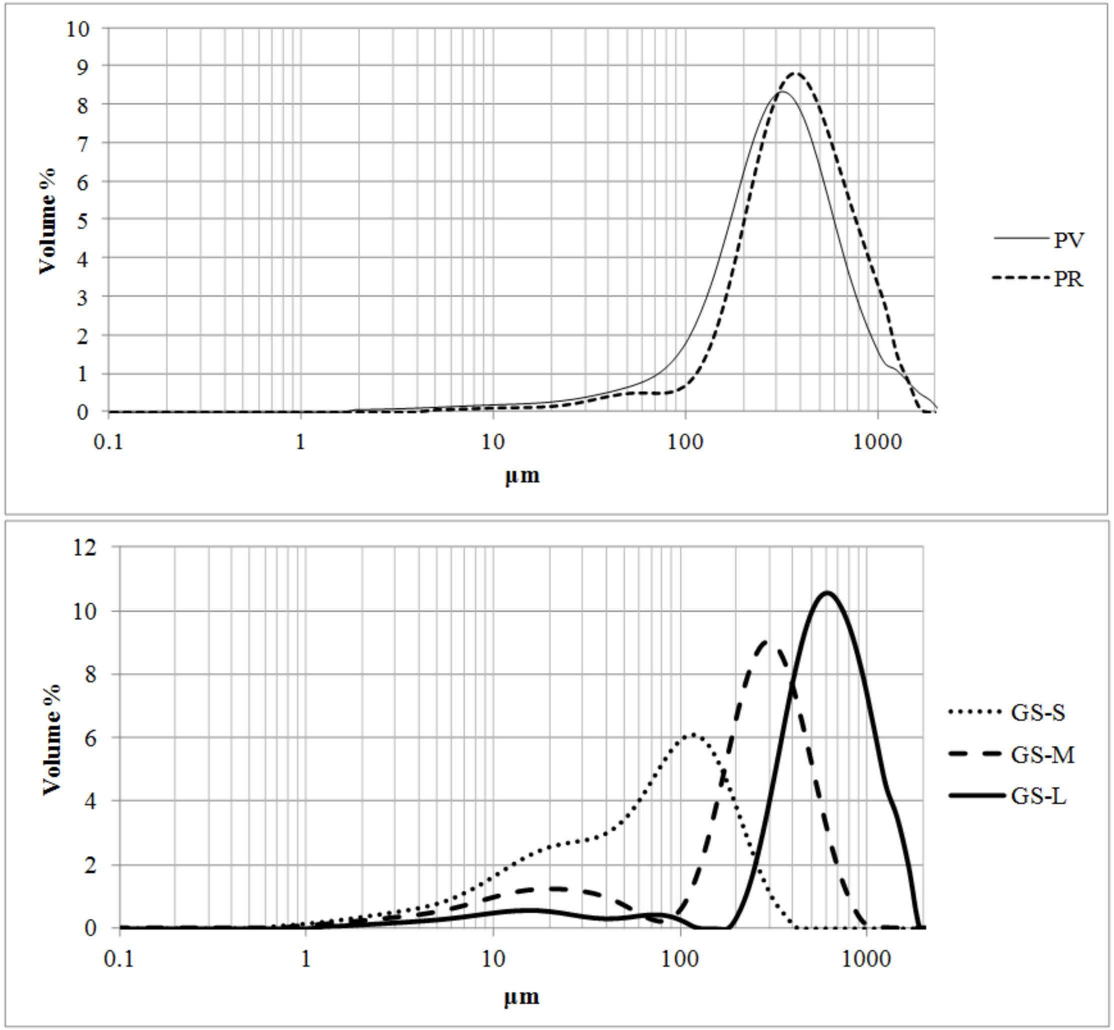

Fig. 2. PSD of PV and PR Tomato Purees and Grape Skins Granulometric Fractions GS-S; GS-M and GS-L. Abbreviations are the Same as in Fig. 1.

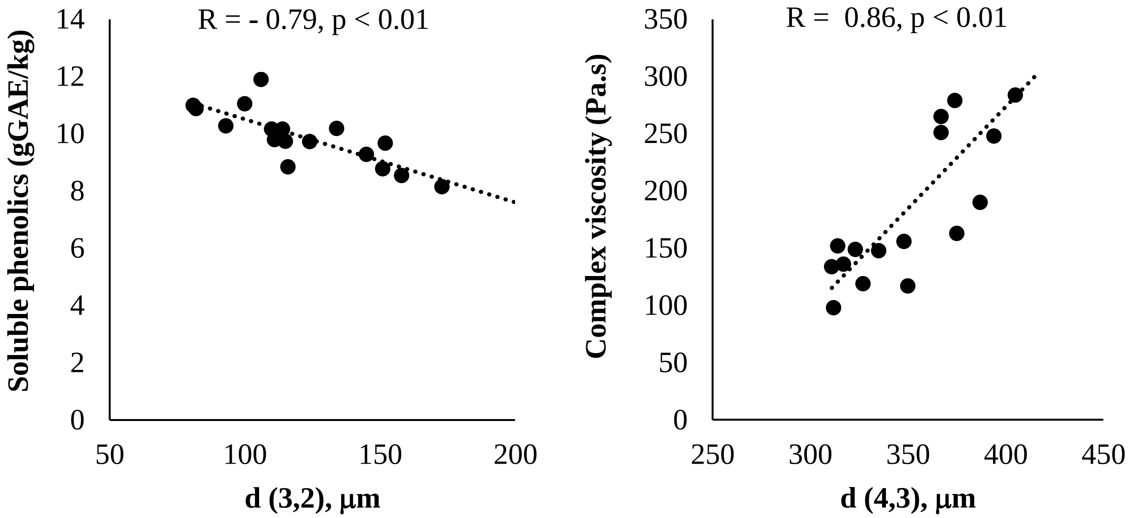

Fig. 3. Correlations between Surface-Weighted Mean Diameter, d(3,2) and Soluble Phenolics (g GAE/kg) and between Volume-Weighted Mean Diameter, d(4,3) and Complex Viscosity for all the Fortified Purees Subjected to Different Treatments (Mixing, Microwave, Autoclave).

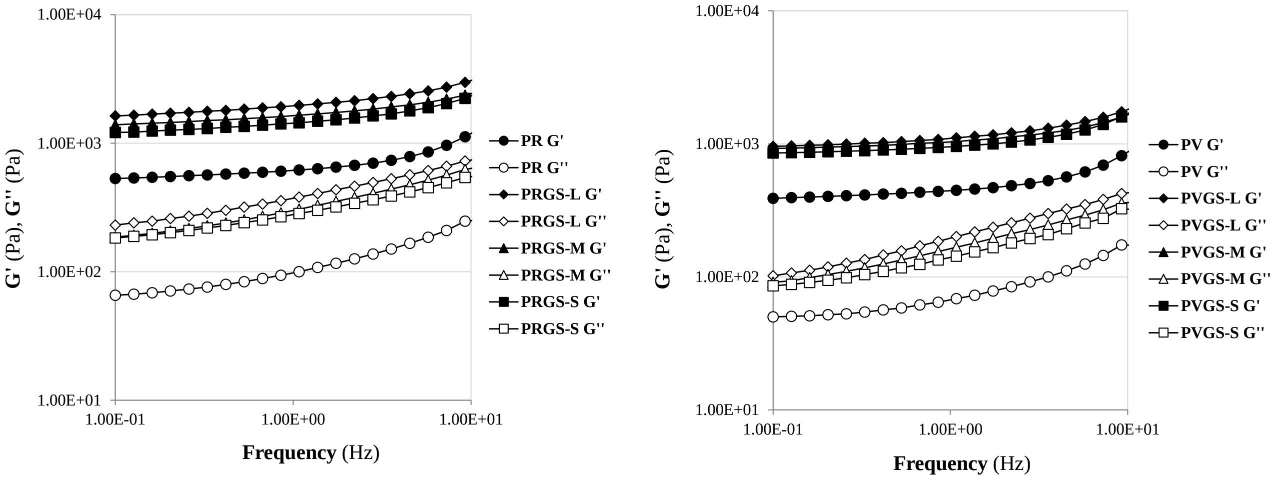

Fig. 4. Frequency Sweep Test: Variation in the Storage Modulus (G’, Pa) and Loss Modulus (G’’, Pa) of Tomato Purees Formulated with GS Fractions and Control Tomato Purees after mixing. Abbreviations are the Same as in Fig.1. (*n* = 3).

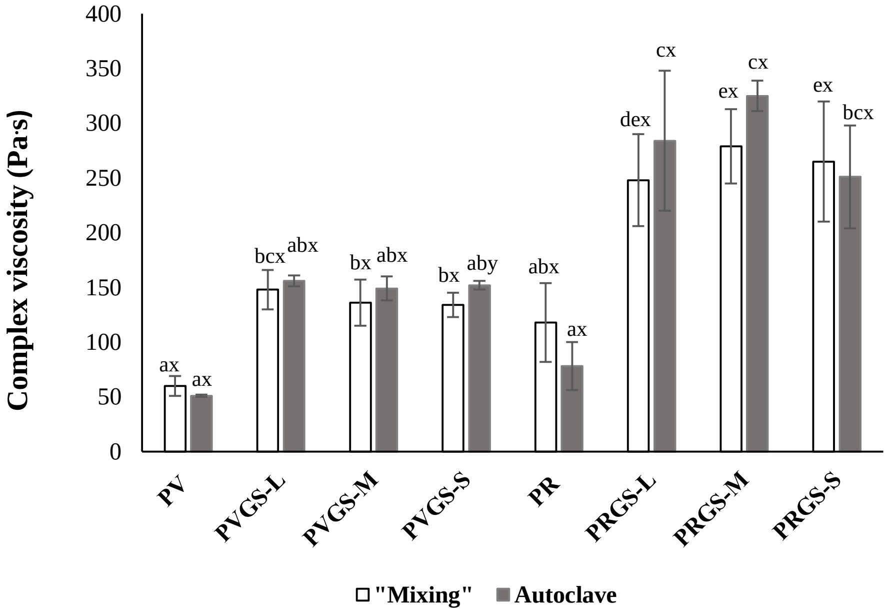

Fig. 5. Complex Viscosity (η*, Pa·s) values (at 1 Hz) of Tomato Purees Formulated with GS Fractions and Control Tomato Purees after Mixing and Autoclave Treatments. Different letters within the same treatment (a –e) or puree (x – y) indicate significant differences (LSD, $p < 0.01$). Abbreviations are the Same as in Fig.1. Error bars indicate SD. (*n* = 3).