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Scattering efficiency of a cloudy apple juice: Effect of particles characteristics and serum composition

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Abstract

The effect of particles and serum characteristics on turbidity of natural, or "regular", and modeled cloudy apple juices was studied in this work. Modeled apple juices were made by re-dispersing a determined quantity of apple particles in a simplified serum (mainly glucose, hydrolyzed pectin, and malic acid in water). Only glucose was found to have a significant effect on turbidity. Particle size was affected by soluble solids concentration, which was attributed to conformational changes in juice particles aggregates, simultaneously with a reduction in particle solvatation. Scattering efficiency was determined in natural and modeled cloudy apple juice both experimentally from a nephelometric method, Q^*_{av}, and theoretically with the Mie theory, Q_{Mie}. Decrease in juice specific turbidity at increasing soluble solids concentrations (X), was governed by the decrease of Q^*_{av} at increasing refractive index of the liquid medium n_m. As predicted by theory, the scattering efficiency increased at increasing particle size, for a constant n_m. Finally, calculated values of Q^*_{av} and Q_{Mie} followed a power law relationship when correlated. This non-linear behavior was explained by considering that Q_{Mie} is the theoretical scattering efficiency of monodisperse-homogeneous spheres, while Q^*_{av} is the experimental nephelometric scattering efficiency of polydisperse-irregular particles.

Keywords: Scattering efficiency; Cloudy apple juice

1. Introduction

Cloudiness in opaque beverages arises from the scattering of light by suspended particles in the continuous phase (Dickinson, 1994; Hernández & Baker, 1991; Hernández, Baker, & Crandall, 1991). Light scattering is the deflection of a light beam by discrete variation in refractive index due to the presence of particles, or by spatial refractive index fluctuations. The extent of light scattered by dispersions is determined mainly by the relationship between the particle size and wavelength, and by the difference in the refractive index between the particles and the surrounding liquid (McClements, 1999).

Cloudy apple juice is a colloidal dispersion where the continuous medium is basically a solution of pectin, sugars and malic acid, and the dispersed matter is mainly formed by cellular tissue comminuted during fruit processing (Benítez, Genovese, & Lozano, 2007; Genovese & Lozano, 2006; Sorrivas, Genovese, & Lozano, 2006). The light scattering of the polydisperse particles in suspension is perceived as juice turbidity (Mollov, Mihalev, Buleva, & Petkanchin, 2006). The literature discusses different models for building a stable cloud particle. The cloud particles contain a core, consisting of protein, which is positively charged. These speculations are supported by those of Yamasaki, Yasui, and Arima (1964) and Endo (1965), in which a carbohydrate shell consisting among others of negatively charged pectin, surrounds particles containing a positively charged protein core. This positively charged core is able to build a complex with negatively charged pectin.

Two methods are generally used to measure turbidity of cloudy beverages: spectrophotometry and nephelometry.

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Spectrophotometers measure the amount of light absorbed by the suspended particles in the beverage, which for dilute systems is often expressed in terms of the optical density (Dickinson, 1994; Hernández et al., 1991):

$$\mathrm{O.D.} = \log(I/I_0) = -\log T_{\mathrm{O}} \tag{1}$$

where I_0 and I are the incident and transmitted light intensity respectively, and T_{O} is the optical transmittance:

$$T_{\mathrm{O}} = \exp(-\tau L) \tag{2}$$

where τ is the turbidity and L is the optical path length (Dickinson, 1994; Hernández & Baker, 1991). It can be observed from Eq. (2) that turbidity obtained by the spectrophotometric method has units of length^{-1}.

Turbidity of dispersions depends on concentration, size, and relative refractive index of their particles (McClements, 1999). For a system of spherical polydisperse particles of volume-surface diameter D_{32}, the turbidity in Eq. (2) is given by Dobbins and Jizmagian (1966):

$$\tau = \frac{3}{2}\frac{C}{\rho_{\mathrm{m}}}\frac{Q_{\mathrm{av}}}{D_{32}} \tag{3}$$

where ρ_{m} is the density of the liquid medium (Hernández & Baker, 1991), and Q_{av} is the average particle scattering efficiency given by:

$$Q_{\mathrm{av}} = \int_0^\infty Q(D)\cdot p(D)\cdot D^2\mathrm{d}D \Big/ \int_0^\infty p(D)\cdot D^2\mathrm{d}D \tag{4}$$

where $p(D)$ is the particle size distribution, and $Q(D)$ is the scattering efficiency of a particle of diameter D (the probability that a photon incident on the particle will be scattered). For the particular case of monodisperse spheres of diameter D, $Q_{\mathrm{av}} = Q(D)$. Scattering efficiency depends on the particle size, the refractive indices of the particle and the continuous phase, and the wavelength of the incident light.

The scattering efficiency is a maximum when the size of the particles is similar to the wavelength of the light impinging into the sample (~0.4–0.7 μm for visible light). This is important for the formulation of food beverages because it implies that the desired turbidity might be obtained at lower particle concentrations, by optimizing the particle diameter to give the maximum amount of scattering. On the other hand, the refractive index of aqueous solutions depends on the type and concentration of solutes present (e.g. sugars, organic acids, and salts). At high concentrations of these components, the refractive index of the particles may be matched to that of the surrounding liquid, which greatly reduces the degree of scattering, causing therefore the dispersion to appear transparent (McClements, 1999).

All the mathematical and computational problems concerning light scattering theory for spherical particles (Mie theory) are well studied and resolved (Michel, 2000). Mie theory is the solution of Maxwell's equations for an isotropic sphere. However, problems still remain to be tackled in the case of non-spherical particles (McClements, 1999; Mishchenko, Travis, & Lacis, 2004).

In the spectrophotometric method, it has been assumed that the only contribution to O.D. is from light scattered by suspended particles. However, for colored beverages there is a contribution also from light absorption, which needs to be eliminated by using colored standards. Another disadvantage of using a spectrophotometer is that it is insensitive to light scattered by small particles ($\leqslant$0.2 μm) (Dickinson, 1994; Hernández & Baker, 1991; Hernández et al., 1991).

For these reasons the opacity of beverages is often determined commercially using nephelometers, which measure the intensity of light scattered, usually at a 90° angle, and compares it with that of a standard suspension. One disadvantage of nephelometric measurements is that they may be disproportionately sensitive to small particles ($\leqslant$0.2 μm), giving turbidity values higher than human's eye can detect ("invisible haze"). This is because nephelometers usually detect light scattered not only forward but also at $90 \pm 30°$ (Dickinson, 1994; Hernández & Baker, 1991; Hernández et al., 1991).

However, this problem was considered to be non significant in cloudy apple juice, where the average particle size is usually >0.6 μm (Benítez et al., 2007; Genovese & Lozano, 2006; Sorrivas et al., 2006). Consequently, turbidity of apple juices studied in this work was measured using a nephelometric (instead of spectrophotometric) method.

As previously explained, Eq. (3) was derived for polydisperse systems of spherical particles in terms of their average volume-surface diameter D_{32}. However, cloudy apple juice is a dilute colloidal dispersion of irregularly shaped, hydrated particles (Benítez et al., 2007; Genovese & Lozano, 2006). The size of this type of particles is better characterized by the hydrodynamic diameter (D_{h}), which is determined as follows.

It is well known that the diffusivity of a particle suspended in a liquid is reduced when the liquid's viscosity increases. For small motion, laminar flow is a reasonable assumption. The diffusion coefficient can be calculated using the Stokes–Einstein equation for dilute ($\ll$1% or so) systems (McClements, 1999):

$$\wp = \frac{k_{\mathrm{B}}T}{3\pi\mu D_{\mathrm{h}}} \tag{5}$$

where $\wp$ is the Brownian diffusion coefficient; k_{B} is Boltzmann's constant; T is the absolute fluid temperature; μ is the dynamic viscosity of the working fluid; and D_{h} is the hydrodynamic diameter, which is generally very close to, but slightly larger than the geometrical diameter of a sphere due to solvatation. The hydrodynamic diameter (D_{h}) is the most appropriate particle size to use in equations relating to fluid-particle interactions; i.e. an equivalent sphere diameter derived from a measurement technique involving hydrodynamic interaction between the particle and fluid. Photon correlation spectroscopy (PCS) is widely used as an analytical tool to measure D_{h} (Xu, 1998).

As the diffusion coefficient is inversely proportional to the size of the particles (Eq. (5)), then, the smaller the particle, the shorter the fluctuation time.

For monodisperse system in Brownian motion, the diffusion of the particles in erratic motion causes the intensity autocorrelation equation to decay exponentially as a function of the linewidth of the spectrum (exp-$t\Gamma$) Berne and Pecora (1976). The spectral linewidth (Γ) is related to the diffusion coefficient by:

$$\Gamma = \wp q^2 \tag{6}$$

where the scattering vector, q, is given by

$$q = (4\pi n_{\rm m}/\lambda_0)\sin(\theta/2) \tag{7}$$

in which $n_{\rm m}$ is the refractive index of the liquid medium, θ is the scattering angle, and λ_0 is the light wavelength in vacuum. By combining Eq. (5) with Eq. (6):

$$D_{\rm h} = \frac{k_{\rm B}Tq^2}{3\pi\mu\Gamma} \tag{8}$$

Photon correlation spectroscopy digitally measures the intensity fluctuations of a signal at the level of the photon. When the logarithm of the autocorrelation function is graphed versus time, the slope of the resulting line is the linewidth (Γ). Using Eq. (8), the size of the scattering particles can then be determined.

The main objective of this work was to obtain information about the effect of particles and serum characteristics on the nephelometric turbidity of both a natural, or "regular", and modeled cloudy apple juices (MCAJ). Modeled apple juices were made by re-dispersing a determined quantity of apple particles in simplified serum (mainly water, sugars and malic acid) solutions. The study includes the effect of medium refractive index and particle size on scattering efficiency. Experimental and Mie theoretical particle scattering efficiency were also compared. Finally a model to predict changes in turbidity was proposed.

2. Materials and methods

2.1. Sample preparation

2.1.1. Regular cloudy apple juice (CAJ)

Regular cloudy apple juice (CAJ) was obtained as follows. Granny Smith apples bought in a local market were milled and pressed in our pilot plant. The juice extracted (12.2 °Brix) was heated in a water bath up to 80 °C and hold during 5 min at that temperature before cooling, to inhibit further enzymatic and microbial activity. Finally, the juice was centrifuged at 2587*g* during different times: 0, 2.5, 5, 10, and 15 min.

Particle concentration, C [g/L], of each sample was determined as follows. A certain volume (≈20 ml) of juice was micro-filtered through a 0.45 μm cellulosic membrane (E04WP04700, MSI, Westboro, MA), such that all the juice particles were retained in the filter paper, which was previously weighed. Finally, the filter with the particles was vacuum dried at 55–60 °C overnight, and weighed. Determinations were made at least in duplicate.

2.1.2. Modeled cloudy apple juices (MCAJ)

In order to obtain modeled cloudy apple juices (MCAJ), a commercial cloudy apple juice, kindly provided by the industry (Jugo S.A., Villa Regina, Rio Negro, Arg.), was enzymatically treated (Solvay 5XLHA; 20 mg/l, 2 h a 50 °C) to eliminate soluble pectin, by following the hot technique (Toribio & Lozano, 1984). Juice depectinization is a mandatory step to reduce serum viscosity and avoid membrane collapse during ultrafiltration. Using this depectinized apple juice, the following samples containing the same concentration of colloidal particles ($C_0 = 0.63$ g/L) were prepared:

1. PAJ: Apple juice clarified by ultrafiltration (permeate).
2. DJ: Diafiltred juice; mainly apple juice particles in water.
3. DJ + P: Diafiltred juice plus hydrolyzed pectin. 0.1 g/L.
4. DJ + MA: Diafiltred juice plus malic acid. 6.33 ± 0.01 g/L.
5. DJ + G: Diafiltred juice plus glucose. 100 g/L.
6. RCAJ: Reconstituted cloudy apple juice (PAJ + particles).

Apple juice was clarified and/or diafiltered (21.4 L water per litter of juice) with an Ultrafiltration system (Osmonic Sepa® CF; Osmonics; Minnetonka; Mn; USA) using a 100 kDa (MWCO) polysulphone membrane. Diafiltration is essentially a dilution process and is performed in conjunction with a concentration process. Water is added while filtrate is removed. Diafiltration was continued until the electric conductivity (ε) remained constant at 0.06 mS/cm.

The particle concentration ($C_0 = 0.63$ g/L) used to prepare samples 2–6 was determined as follows. Diafiltered juice (25 mL) was freeze-dried in a HETO Model FD 8.0 (Heto-Holten, Denmark) freeze dryer. The heating plate temperature was set to 20 °C and the vacuum to 0.1 mBar to initiate drying. After drying for 48 h, apple juice particles were removed from trays, stored under vacuum in desiccators with P_2O_5 and weighed to determine the original particle concentration of the diafiltered juice. Then, a 3.3 ml aliquot of the diafiltered juice was diluted to 15 ml with the proper solution (pectin, malic acid, glucose, water or permeate) to obtain the sample with $C = C_0$. Samples with lower particles concentrations (0.12–0.63 g/L) were obtained by further dilution with the corresponding solution.

Malic acid content was determined by titration in accordance with the method reported by the IFFJP, (1974). Initial malic acid content resulted 6.33 ± 0.01 g/L. All chemicals were analytical grade from Sigma Chem. Co. (St. Louis, MO).

2.2. Measurements

Soluble solids concentration, X [°Brix], and relative refractive index of the liquid medium, n_m, were measured in a bench refractometer (AO Scientific, Abbe Mark II, Reichert, USA) at 20(±0.1) °C.

Nephelometric turbidity (τ_n) was measured in a PC Compact Turbidimeter (Aqualitic, Germany) in nephelos turbidity units (NTU), with an incident infrared light of 875 nm and a scattering angle of 90°. Samples of the juice were placed in a 15 ml cell, capped, and gently inverted twice to ensure even mixing. Secondary standards of 100 or 1000 NTU were used, depending on the sample.

Particle size distribution, average hydrodynamic diameter (D_h), and electric conductivity (ε) were determined with a Malvern Zetasizer 3000 (Malvern Instruments Inc., London). The equipment requires the viscosity of the dispersant for calculations (Eq. (8)), which was obtained from tables for glucose solutions (Wolf, Brown, & Prentiss, 1987). All measurements were done at least in triplicate. In all cases, coefficients of variation were less than 10%.

2.3. Refractive index of particles

If a particle has the same refractive index as the media surrounding it, then the particle will disappear. Therefore, refraction of light is minimum under that condition (Taisne, Walstra, & Cabane, 1996). Consequently, the methods employed to determine the refractive index of particles in a suspension usually consist in immersing particles in liquids of various refractive indices. One method is to determine when the specific turbidity reaches a minimum (zero), as observed in an oil in sucrose solution emulsion (McClements, 1999). Another method is to observe in a microscope when the Becke's lines disappear (Becke's line test). In this case, freeze dried apple particles were re-suspended in sucrose solutions in the range 53–75 °Brix, and the behavior of Becke's lines was observed in a Nikon Model Labophot 2 microscope with a 100 × 1.25 immersion objective. Similarly to emulsions only the edge of apple particles, and not the entire particle as in the case of hard spheres, disappear. By convention, the microscope is defocused by increasing distance between objective and sample.

3. Results and discussion

As expected, nephelometric turbidity (τ_n), particle concentration (C) and average hydrodynamic diameter (D_h) of regular cloudy apple juice (CAJ) decreased at increasing centrifuging times (Table 1) due to sedimentation of the coarse-unstable particles. Values of τ_n were relatively low (<600 NTU) as compared with other studies (Mollov et al., 2006), which were attributed to the variety and degree of ripeness of the apples used to make the juice. Refractive index of the liquid medium was determined to be $n_m = 1.351$, and its density ($\rho_m = 1.053$ g/ml) was obtained from literature (Constenla, Lozano, & Crapiste, 1989).

Table 1
Nephelometric turbidity (τ_n), particle concentration (C) and mean hydrodynamic diameter (D_h) of regular cloudy apple juice (CAJ) as a function of centrifuging time (t) at 2587g

t (min)	τ_n (NTU)	D_h (μm)	C (g/L)
0	551 ± 2	1.01 ± 0.01	2.61 ± 0.14
2.5	308 ± 4	0.65 ± 0.01	2.25 ± 0.22
5	217 ± 1	0.54 ± 0.00	2.03 ± 0.00
10	192 ± 1	0.44 ± 0.00	1.94 ± 0.28
15	177 ± 1	0.43 ± 0.00	1.88 ± 0.07

3.1. Effect of soluble solids on particle size

The addition of glucose to a diafiltered juice (DJ + G) showed that particle size was affected by soluble solids concentration. Fig. 1 shows the decrease of D_h [μm] with increasing weight percent of glucose, X_G [% w/w]. Since experimental D_h values were not affected by particle concentration, C, in the range 0.12–0.63 g/L (results not shown), experimental data was fitted by a quadratic polynomial ($R^2 = 0.990$):

$$D_h = 1.05 - 1.32 \times 10^{-2} X_G + 5.82 \times 10^{-5} X_G^2 \tag{9}$$

Eq. (9) is valid for X_G in the range 0–60% w/w.

It has been proposed (Hiemenz, 1986) that in colloidal systems where neither aggregation nor segregation of particles occurs, radius, or gyro-radius which is the radius of the circle made by a gyrating particle, can be reduced by modification of the solvatation, or the sphericity. Size

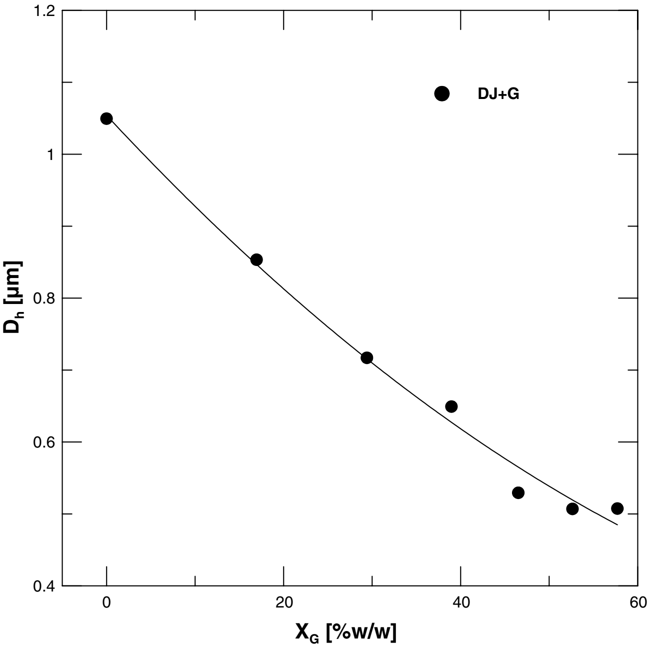

Fig. 1. Effect of glucose concentration (X_G) on the particle hydrodynamic diameter (D_h) of diafiltered juice (DJ + G). Solid line represents Eq. (9).

reduction observed in Fig. 1 may be attributed to conformational changes in juice particles aggregates, from an extended to a more spherical conformation. Moreover, and simultaneously, as glucose can fit water reducing water activity a_w (Sato, Kawabuchi, Irimoto, & Miyawaki, 2004), a reduction in particle solvatation with higher glucose concentration may be expected.

It should be also noted in Table 1 and Fig. 1, that values of D_h were >0.4 μm, indicating that the presence of very small particles affecting nephelometric turbidity measurements may be discarded. Furthermore, the particle size range was similar to the wavelength of visible light (~0.4–0.7 μm), which is known to maximize the turbidity for a given particle concentration.

3.2. Effect of particles and serum characteristics on nephelometric turbidity

Fig. 2 shows the effect of malic acid, glucose, and hydrolyzed pectin on nephelometric turbidity, τ_n, of a diafiltered apple juice (DJ) with constant particle concentration, as compared with a reconstituted apple juice (RCAJ). It can be observed that the turbidity of the DJ was higher than the RCAJ. However, the addition of glucose to the diafiltered juice (DJ + G) drastically reduced the turbidity to values as low as those of the RCAJ. No significant effects were observed when malic acid (MA), or hydrolyzed pectin (P), was incorporated into the diafiltered juice. From the several factors that may affect turbidity (Eq. (3)), it will be demonstrated in the next section that the lower turbidity of the juices with sugar (RCAJ and DJ + G) was mainly caused by their higher refractive index of the medium (n_m), which reduced the average scattering efficiency (Q_{av}) of their particles.

This behavior is observed in more detail in Fig. 3, which shows the effect of soluble solids content on turbidity of RCAJ and DJ + G, in the range 0–10% w/w (or °Brix), keeping constant the concentration of particles ($C_0 = 0.63$ g/L). Results indicated that independently of the component in solution (only glucose; or glucose, fructose, malic acid, hydrolyzed pectin, polyphenols and the other components of a clarified apple juice) nephelometric turbidity decreased with soluble solids by following the same behavior.

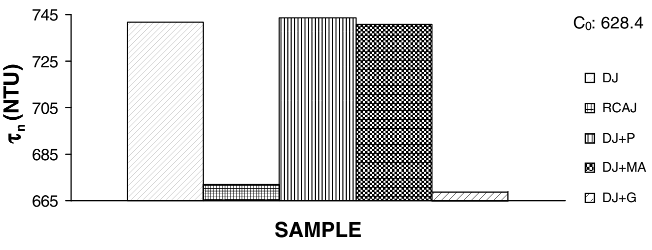

Fig. 2. Effect of malic acid (MA), glucose (G), and hydrolyzed pectin (P) on the nephelometric turbidity of a diafiltred apple juice (DJ) with constant particle concentration, as compared with a reconstituted apple juice (RCAJ).

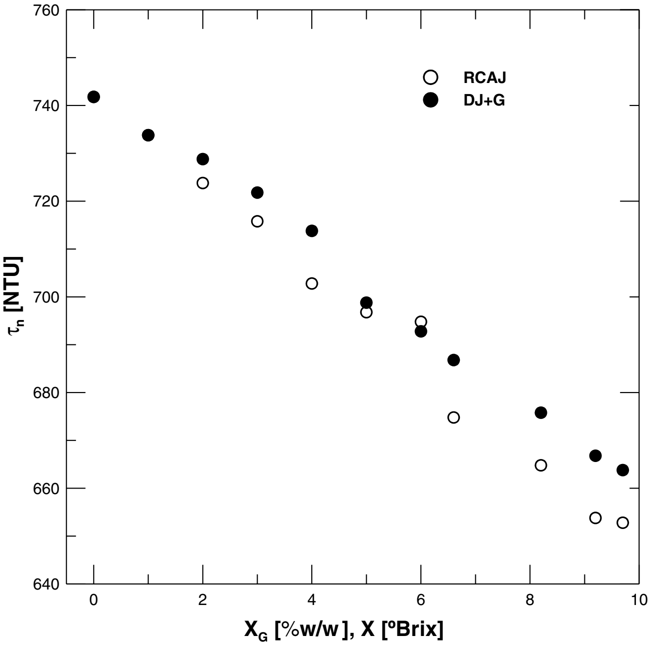

Fig. 3. Effect of glucose concentration (X_G) and soluble solids content (X) on the nephelometric turbidity of a diafiltred juice (DJ + G) and a reconstituted juice (RCAJ), respectively, with constant concentration of particles ($C_0 = 0.63$ g/L).

The effect of particle concentration, C, on turbidity of a diafiltred juice with different glucose concentrations (DJ + G) is shown in Fig. 4. In accordance with theoretical Eq. (3), juice turbidity was found to be directly

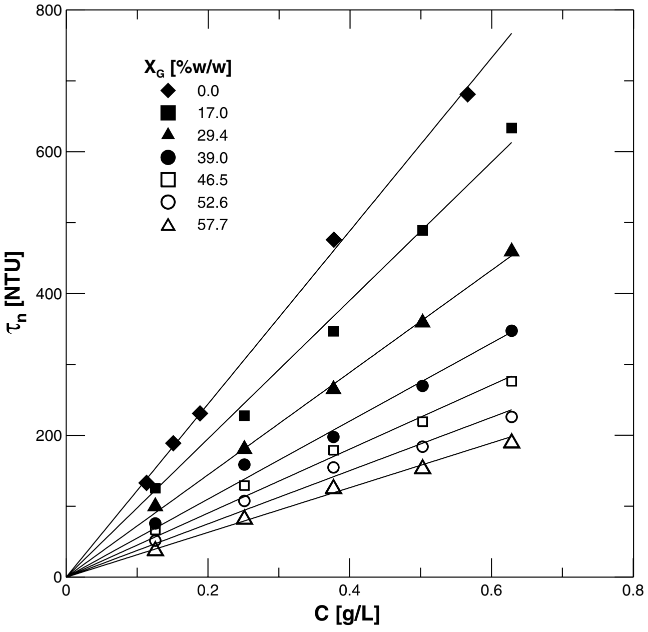

Fig. 4. Effect of particle concentration (C) on nephelometric turbidity of diafiltered juice with different glucose concentrations (X_G). Experimental data (symbols) fitted with Eq. (10) (solid lines).

proportional to particle concentration. Consequently experimental data were fitted with straight lines-through-origin:

$$\tau_n = bC \tag{10}$$

where b is known as the specific turbidity (Dickinson, 1994). It was obtained from the slope of each line, and summarized in Table 2. It can be observed that the specific turbidity decreased at increasing glucose concentrations (X_G). This is equivalent to the decrease of τ_n (for a constant C) at increasing X or X_G, observed in Fig. 3. Again, this behavior is governed by the decrease of Q_{av} at increasing n_m, as demonstrated in the next section.

3.3. Effect of medium refractive index and particle size on scattering efficiency

As previously mentioned, the average hydrodynamic diameter (D_h) and the nephelometric turbidity (τ_n) are easy to measure, widely used parameters to describe the characteristic particle size and turbidity of commercial beverages. In this sense, they are more appropriate than the volume-surface diameter (D_{32}) and the spectrophotometric turbidity (τ), respectively. Consequently, in order to obtain a practical and probably more suitable expression of Eq. (3) for cloudy apple juice and similar dispersions, D_{32} was replaced by D_h, and τ was replaced by τ_n. After combination with Eq. (10), a modified average scattering efficiency was obtained:

$$Q^*_{av} = \frac{2}{3} b \rho_m D_h \tag{11}$$

From a dimensional analysis, values of Q^*_{av} calculated in this way have units of [NTU m]. These values represent the nephelometric average scattering efficiency of the polydisperse, irregular juice particles with average diameter D_h.

In the first place, values of Q^*_{av} were calculated for diafiltered juice with different glucose concentrations (DJ + G), using Eq. (11). While values of b were obtained from Table 2; values of D_h and ρ_m resulted from the experimental data shown on Fig. 1 and tables for glucose solutions (Wolf et al., 1987), respectively. Calculated Q^*_{av} values were plotted as a function of the refractive index of the liquid medium, n_m (Fig. 5). Values of n_m were also obtained form tables for glucose solutions (Wolf et al., 1987). Within this experimental range, Q^*_{av} values decreased at increasing n_m values. Theoretically, when the liquid medium has the same refractive index of the particles, that is $n_m = n_p$, the scattering of light is minimum (zero), but increases as n_m moves to higher or lower values (McClements, 1999). Consequently, experimental data were fitted with a parabola (Eq. (12)) such that at $n_m = n_p$, it accomplishes both theoretical conditions: $Q^*_{av} = 0$, and $\mathrm{d}Q^*_{av}/\mathrm{d}n_m = 0$:

$$Q^*_{av} = m(n_p - n_m)^2 \tag{12}$$

where m is an adjustable constant. Resulting fitting parameters were $m = 35.55$, $n_p = 1.487$, and $R^2 = 0.994$.

Table 2
Specific turbidity (b) as a function of glucose concentration (X_G), obtained by fitting Eq. (10) to experimental data in Fig. 4

X_G	b (NTU L/g)	R^2
0.0	1221	1.000
17.0	976	0.999
29.4	722	1.000
39.0	550	0.998
46.5	451	0.997
52.6	376	0.996
57.7	316	0.998

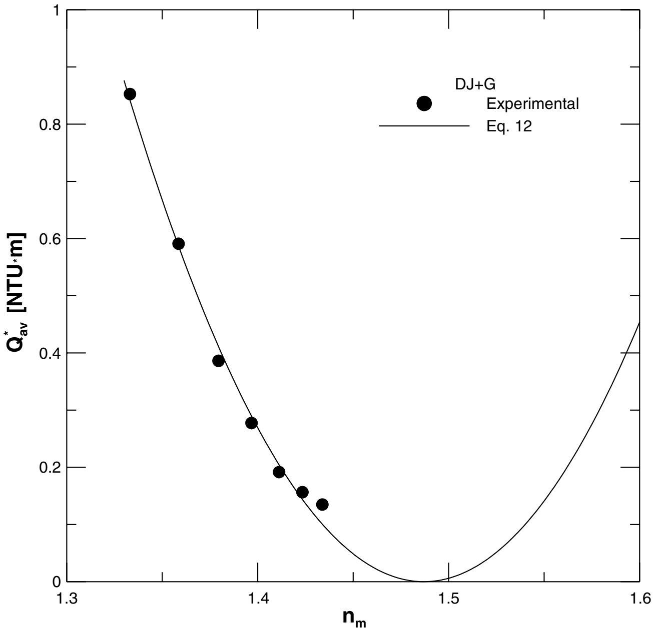

Fig. 5. Effect of the refractive index of the medium on the modified average scattering efficiency of apple juice particles, calculated from Eq. (11). Solid line represents Eq. (12).

Coincidently, the Becke lines method indicated that similar refractive index between apple particles and sucrose solution was observed in the range of 65–70 °Brix, equivalent to $n_p = 1.453$–1.465. In the literature the refractive index of adsorbed protein layers is usually preset and its assumed value varies between 1.35 and 1.6 (Benesch, Askendal, & Tengvall, 2002; Jung, Campbell, Chinowsky, Mar, & Yee, 1998). For the refractive indices of protein layers realistic values between 1.36 and 1.55 were obtained depending on size of the proteins and the experimental conditions (Vörös, 2004). On the other hand, the refractive index of the other major components of apple tissue, cellulose and pectin, are $n = 1.56$ and $n = 1.504$, respectively (Don, 1991; McCrone & Delly, 1992).

Since the refractive index of the liquid medium, n_m, is proportional to the soluble solids concentrations (X or X_G), it follows that Q^*_{av} decreased at increasing X_G in the experimental range studied in this work (0–60% w/w). This result will help us to explain the behavior observed in Fig. 3 and Table 2. From Eq. (11), it follows that the decrease of specific turbidity ($b = \tau/C$) at increasing X_G should be produced by a decrease in the product $(Q^*_{av}/\rho_m \cdot D_h)$. It was calculated that the denominator $(\rho_m \cdot D_h)$ decreased at increasing X_G from 0% to 60% w/w, which means that the decrease of b was produced by the decrease of Q^*_{av}. In other words, the decrease of specific turbidity at increasing soluble solids concentration was governed by the decrease of Q^*_{av} at increasing n_m in the experimental range studied in this work (1.33–1.43).

In second place, values of Q^*_{av} were calculated from the experimental data shown on Table 1 for regular cloudy apple juice (CAJ) with different particle sizes and concentrations, using Eqs. (10) and (11). Calculated Q^*_{av} values were plotted as a function of the average hydrodynamic diameter, D_h (Fig. 6). As predicted by theory, the scattering efficiency increased at increasing particle size, for a constant refractive index of the liquid medium.

3.4. Correlation between experimental and theoretical scattering efficiency

In order to confirm that experimental Q^*_{av} values obtained with Eq. (11) are in accordance with theoretical predictions, values of the scattering efficiency of homogeneous spheres of diameter D_h were calculated with the Mie Theory, Q_{Mie}, using a computer program available on Internet (Michel, 2000). Calculated Q_{Mie} values are dimensionless. Parameters used for calculations were:

(a) The real part of the refractive index of the particles: $n_p = 1.487$, obtained in this work with the turbidity method;
(b) The imaginary part of the refractive index of the particles $n'_p = 0.01$, reported for milk (Meyer et al., 1998);
(c) The refractive index of the liquid medium: n_m, determined for each sample; and
(d) The vacuum wavelength of the incident radiation: $\lambda = 875$ nm, the operative wavelength of the turbidimeter used in this work.

Calculated values of Q^*_{av} and Q_{Mie} were correlated in a double logarithmic plot (Fig. 7) for both regular cloudy apple juice (CAJ), and diafiltered juice with glucose (DJ + G). It is clear that Q^*_{av} and Q_{Mie} followed a power law relationship in both juices. Consequently, experimental data was fitted with the expression:

$$Q^*_{av} = \alpha \cdot Q^{\beta}_{Mie} \tag{13}$$

where α and β are fitting parameters, summarized in Table 3 for CAJ and DJ + G, with highly satisfactory regression coefficients ($R^2 > 0.99$). This result indicate that Q^*_{av} trends are consistent with theory. It was assumed that Q^*_{av} and Q_{Mie} did not follow a linear relationship because they do not have exactly the same physical meaning: Q_{Mie} is the theoretical scattering efficiency of monodisperse-homogeneous spheres, while Q^*_{av} is the experimental nephelometric scattering efficiency of polydisperse-irregular particles. Consequently, parameters α and β in Eq. (13) may be associated with particles size distribution $p(D)$ and shape.

This hypothesis was qualitatively supported by comparing the particle size distributions of regular cloudy (CAJ)

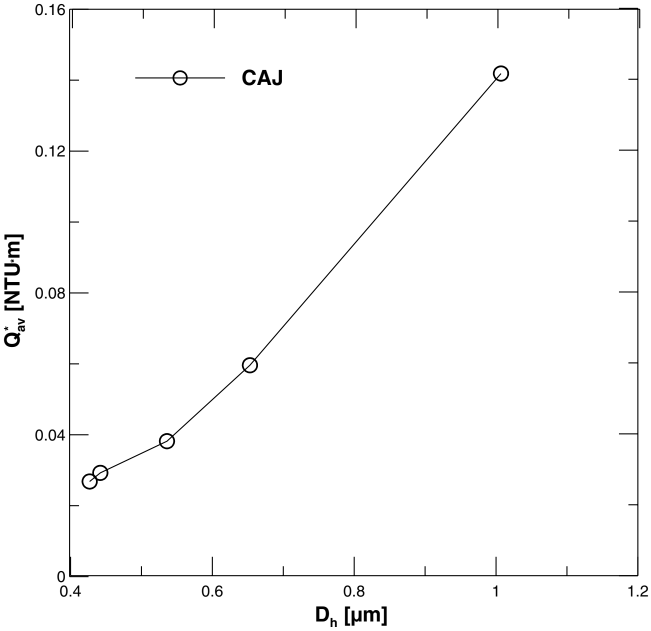

Fig. 6. Effect of the average hydrodynamic diameter on the modified average scattering efficiency of apple juice particles. Calculated using experimental data from Table 1 in Eqs. (10) and (11).

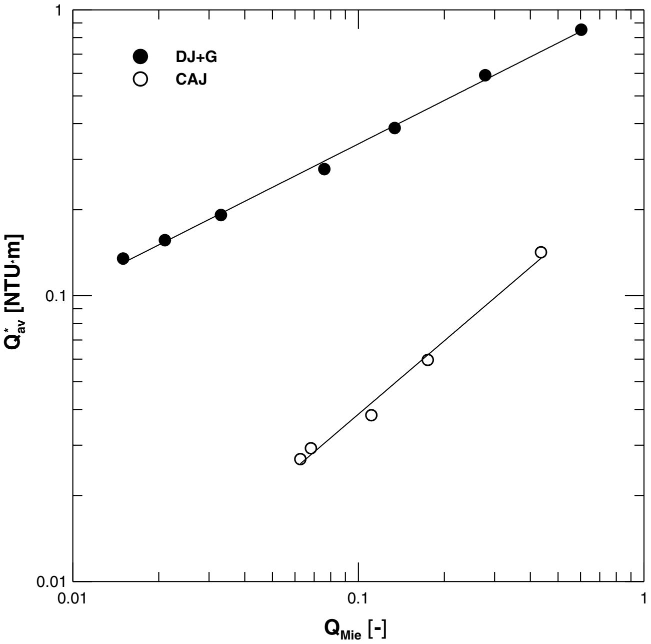

Fig. 7. Correlation between the modified average scattering efficiency (Q_{av}) of apple juice particles, and Mie's scattering efficiency (Q_{Mie}) of monodisperse spheres. Experimental data on regular cloudy apple juice (○), and diafiltered juice with glucose (●), fitted with Eq. (13) (solid lines).

Table 3
Fitting parameters of Eq. (13) for regular cloudy apple juice (CAJ) and diafiltered juice with glucose (DJ + G)

Sample	α (NTU m)	β (–)	R^2 (–)
CAJ	0.275	0.853	0.991
DJ + G	1.087	0.505	0.997

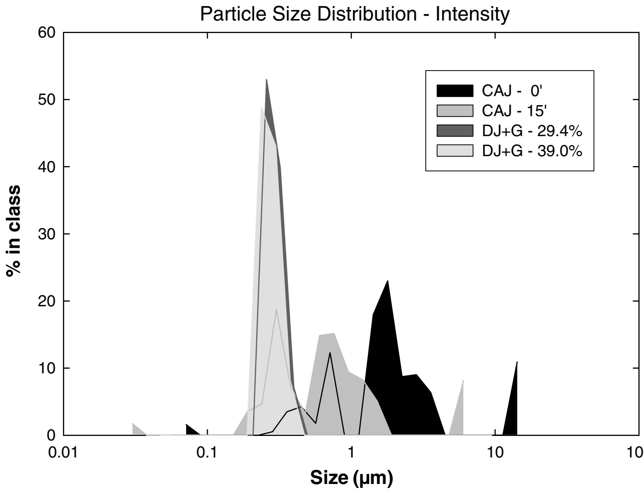

Fig. 8. Particle size distributions of diafiltered juice with glucose (DJ + G) at two different concentrations, and regular cloudy apple juice (CAJ) at two different centrifuging times.

and diafiltered, with glucose, (DJ + G) juices (Fig. 8). CAJ showed particle size distributions more spread than (DJ + G). This result, in addition to probable morphological differences between particles due to different juice processing, may explain the differences in their average scattering efficiencies (Fig. 7).

4. Conclusions

The method proposed in this work for the determination of the scattering efficiency from nephelometric turbidity and hydrodynamic diameter data, was found to be consistent with the Mie Theory. Further studies are required to quantitatively determine the effect of particles size distribution and shape.

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