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Cellulose acetate membrane filtration effect on particle size distribution of golden delicious apple juice: Experimental validation of a simulation model

Efecto de la filtración por membranas de acetato de celulosa en la distribución de jugo de manzana golden delicious: Validación experimental de un modelo de simulación

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Abstract
Juice filtration through membranes is becoming an important technology due to its capacity to keep high quality and sensorial characteristics. In the process, the particle size of the feed and the pore size of the membrane give important information about the characteristics and stabilization of the filtered juice. In this work we present the validation of a previously reported novel computational model used to describe two apple juice filtration systems. Experimental data on the filtration of Golden Delicious (GD) apple juice after two different pre-filtration treatments was obtained and the effect on particle size distribution was studied. The experimental data was compared to the simulation results in which the final mean particle size of filtered juices is determined knowing the pore size distribution of the membrane and the feed particle size distribution. According to the simulated values, the model is valid as a first attempt to provide criteria for membrane selection in the filtration of apple juice as there was a good correlation of the process with the model parameters. The results also show that simulation modeling and analysis are important emerging tools that can be used throughout the process industries including food processing.
Keywords: Apple juice, filtration, membrane, model, validation.

Resumen
La filtración de jugos con membranas es una tecnología importante ya que mantiene una alta calidad y características sensoriales. El tamaño de partícula de la corriente de alimentación y el tamaño de poro de la membrana brindan información importante sobre las características y estabilización del jugo filtrado. En este trabajo presentamos la validación de un modelo computacional reportado previamente. Se describen dos sistemas de filtración de jugo de manzana Golden Delicious (GD) que siguieron dos tratamientos de prefiltración, se estudió el efecto sobre la distribución del tamaño de partícula. Los datos experimentales se compararon con los resultados de una simulación en la que el tamaño medio final de las partículas de los jugos filtrados se determina mediante la distribución del tamaño de los poros de la membrana y la distribución del tamaño de las partículas de la alimentación. Con base a los valores de la simulación, el modelo es válido para proporcionar criterios preliminares de selección de membranas en la filtración de jugo de manzana ya que existe buena correlación del proceso con los parámetros del modelo. Los resultados muestran que las simulaciones y análisis son herramientas emergentes importantes que pueden utilizarse en la industria incluyendo el procesamiento de alimentos.
Palabras clave: jugo de manzana, filtración, membrana, modelo, validación.

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1 Introduction

Changes in lifestyle, concern for fitness, and the need for a balanced diet and new food formulations have resulted in a growing consumption of healthy food products like fruit and vegetables juices (Chatterjee *et al*., 2020, Santacruz-Vázquez *et al*., 2020). In order to expand the juice market, manufacturers are introducing preservative-free and sugar-free fruit juices. In 2021 the global fruit juice market reached 141 Billion US$ (IMARC group, 2022) and for 2027 a compound annual growth rate of 4.31% is expected due to changes in consumer needs during and after the pandemic. Currently, consumers are more aware of their personal health and there is a trend to change carbonated drinks for juices. Fruit juices are heterogeneous suspensions of particles that are mainly composed by cellular tissue fragments (produced during fruit grinding) dispersed throughout an aqueous solution of sugars, ions and other soluble compounds (Dahdouh *et al*., 2016). The particles in fruit juices represent a challenge for long term stability resulting in undesirable characteristics (Genovese and Lozano, 2000). Due to this reason, many processes for size reduction or elimination of particles are carried out during juice production. These processes can involve centrifugation, filtration, homogenization and even the use of biocatalysts to keep only colloidal particles (Carbonell *et al*., 2011, Nawaz *et al*., 2021).

Given the right process conditions, filtration through membranes is becoming an important technology in food science due to its capacity to keep high quality, sensory and nutritional properties (Conidi *et al*., 2020). It has been demonstrated that membrane filtration has a positive effect in avoiding browning and turbidity of apple juice (Fitri and Widiastuti 2017; Gulec *et al*. 2017). But, on the other hand, excessive filtration can cause the loss of flavor and lower nutritional quality. There is an increasing trend in adding functional ingredients to beverages such as fibers, hydrocolloid stabilizers and prebiotics (Beristain *et al*., 2020, Santacruz-Vazquez *et al*., 2020). Therefore, the selection of a proper membrane is a crucial point in juice and beverages processing. Biopolymers are some of the potentially explored materials for membrane separation. One of the most commonly used polymers is cellulose acetate (CA); membranes of this polymer have high flux (Peng and Masanao, 2012) resulting in a good quality juice in which both; browning and turbidity are controlled (Fitri and Widiastuti, 2017).

There are two important characteristics to consider in membrane filtration processes: 1) the particle size distribution of the suspended solids of the feed and 2) the pore size distribution of the membrane. It has been demonstrated that the particle size of the feed and the pore size of the membrane could give important information about the quality and stabilization of filtered juice and are also related to membrane fouling (Fitri and Widiastuti, 2017).

Since the particle size of juices is affected by all the pre-filtration and filtration processes (Stinco *et al*., 2012) and as there are many types of membranes with a wide range of pore sizes, a wide window of exploration (combination of particle and pore sizes) is available to choose the best size reduction and filtration process. Thus, as in many science and engineering areas of research, alternative methodologies like computational simulation and modeling have been employed as low-cost exploration tools for a better understanding of systems. Mathematical modeling is a useful tool to design, control and optimize various processes (Sánchez-Vargas and Valdés-Parada, 2021). Models can help to explore and better understand processes by studying the correlation between input and output parameters; however, their validation is necessary in order to determine their accuracy in predicting real processes when comparing them to experimental data.

Recently, a novel membrane filtration simulation methodology was described. This model predicts the final particle size distribution after a filtration process using both; information of the initial particle size distribution and of the membrane pore size distribution. Specifically, these size distributions are defined by two parameters, the mean, μ (μm), and the standard deviation, σ (μm), of the corresponding Gaussian distribution of the common logarithm of the diameter. The methodology was proposed for a hard sphere model of a colloidal fluid and a membrane composed of a set of parallel planes with circular pores (Marrufo-Hernández *et al*., 2018). There are two remarks to make about the aforementioned simulation model. First, its purpose was to determine empirical correlations between six variables describing the size distributions of the suspended solids in the feed (μ_s, σ_s), the pores of the membrane (μ_p, σ_p) and the solids in the filtrate (μ_f, σ_f). Empirical correlations of this type, as those reported by Marrufo-Hernández *et al*., 2018, allow to predict the size distribution of the filtrate, once the size distributions of the feed and of the pores have been specified. Nevertheless, it is not the only way in which those relations could be useful. It is possible to employ them in membrane selection, since the size distribution parameters of the pores can be determined from the knowledge of the feed and the desired filtrate size distributions. From a more technological perspective, theoretical predictions could contribute to savings in time and resources in the optimization of a juice filtration process in industry, since this can significantly reduce the number of experiments needed to define the operating conditions parameters.

Second, like many other theoretical models on colloidal physical chemistry, the first approximation considers only excluded volume effects. Thus, the application of such model to predict properties of real systems gives valuable information about the role of the geometrical constraints in the phenomenology of the system under study. The model only considers suspended solids in fruit juice as virtually spherical with a well-defined log-normal size distribution as obtained from experimental data. Other interactions such as direct particle-particle (electrical, van der Waals, etc.) and mediated by the solvent (electrical double layer, depletion forces, etc.) (Benítez *et al*., 2009, Dahdouh *et al*., 2016) were not considered. Under some conditions, these interactions could cause particle aggregation with a resulting modification in size distribution. On the other hand, particle-membrane interactions that are determinant in the membrane fouling, resulting in an effective reduction of the pore sizes were not considered either. Even though this model is an idealization of a real filtration process, it was of interest to validate it using systems of significant complexity such as fruit juices that contain colloidal particles of pectin and xylan, proteins and condensed tannins that can contribute to cloudiness (Beveridge, 2002, Hassan *et al*., 2020). In order to do this, data of filtration of GD (Golden Delicious) apple juice using two different pre-filtration treatments (sedimentation and centrifugation) and CA membranes with two different pore sizes was used in the model proposed by Marrufo-Hernández *et al*. (2018), to observe the effect on the size distribution of the suspended particles in the filtered juice.

2 Materials and methods

2.1 *Juice extraction and pre-filtration treatment*

Ripe GD apples from the Cuahutémoc region of Chihuahua (the state that produces 83% of the total apple production in Mexico) (SAGARPA, 2021) were purchased from a supermarket. The apples had reached commercial maturity, and were a good representation of the fruit that is available all throughout Mexico. In a typical experiment, nine to ten apples of approximately 100g from a batch of thirteen were washed, cored and cut in four pieces prior to juice extraction of the unpeeled fruit with a Turmix juice extractor to produce 800 mL of juice. This was followed by enzymatic inactivation achieved by heating at 80°C during 5 minutes as reported (Marrufo-Hernández *et al*., 2017). Two different pre-filtration processes were carried out 1) sedimentation: the unstable suspended particles in the juice were allowed to precipitate by gravity during 30 min, 500 mL of the supernatant was collected and 250 mL were used as sedimented juice (SJ) and 2) centrifugation: centrifuged juice was prepared using the same procedure as SJ followed by a centrifugation to reduce the particle size. For this step 350 mL of juice after sedimentation were centrifuged at 4200 x g for 10 minutes in an Eppendorf 5810R centrifuge at room temperature, 250 mL of the supernatant were used as centrifuged juice (CJ) (Genovese *et al*., 1997).

2.2 *Filtration process*

Apple juice (SJ or CJ) was filtered using a syringe with CA membranes (M1 and M2) Grace (Cat. 62131) and Titán (Cat. A2SRI00064) with two different pore sizes mean pore diameters (D) D_{M1}=0.865 μm and D_{M2}=1.07 μm. The filtrate (60 mL) was recovered and characterized (section 2.3). Filtered samples were labeled with the following nomenclature SJ-M1, CJ-M1, SJ-M2, CJ-M2 where M1 and M2 correspond to the two membranes used to filter the apple juice.

2.3 *Apple juice characterization*

All juices (pre-treated and filtered) were characterized as follows: pH was measured with a multiparameter device HANNA HI98130 series using 20 mL of juice. Sugar content and refractive index (RI) were measured using a volume of 0.1 mL of juice with an ATAGO Abbe NAR-1T LIQUID refractometer, all measurements were made by triplicate prior or after filtration.

All juices (pre-treated and filtered) were characterized by Dynamic Light Scattering (DLS) and Scanning Electron Microscopy (SEM) as follows: For each DLS experiment, 3 mL of juice (prior or after filtration) were poured in a sample plastic cuvette and degassed using a Thermovac degasser. Particle size distributions were measured in a Malvern Zetasizer nano ZS ZEN- model 3500 at 22°C and scattering angle of 173°. The light scattering results were analyzed in the Zetasizer software version 7.03. All experiments were performed in triplicate.

Microscopy analysis of the juice particles was performed in a JSM 5900 LV (Jeol) Scanning Electron Microscope (SEM). Pre-treated juices (SJ and CJ) and filtered juices (SJ-M1, SJ-M2, CJ-M1 and CJ-M2) were dried prior to the analysis. Once dry, the samples were covered with gold with a Denton Vacuum, LLC sputter coater. Magnifications ranged from 100x to 10,000x with an operating voltage of 13 to 15 kV. Images were obtained with the secondary electrons (SE) signal (Genovese and Lozano, 2000).

2.4 Membrane characterization

Membranes before and after the filtration process were characterized using a JSM 5900 LV (Jeol) Scanning Electron Microscope. Samples were mounted on an aluminum sample holder and sputtered coated with gold (Denton Vacuum, LLC). Magnifications ranged from 100x to 10,000x and the operating voltage from 13 to 15 kV. Images from secondary electrons (SE) of the pristine/used membranes were analyzed to identify pores and their shape. To obtain the pore size distribution, the diameter of the pores in each image was measured with the size scale using the image J software version 1.32 (Zhao *et al*., 2000).

2.5 Computational study of the filtration system

The filtration experiment performed in this work was modeled by a computer simulation technique reported recently (Marrufo-Hernández *et al*., 2018). Briefly, the numerical experiment consists of a Brownian dynamics simulation of a colloidal fluid modeled as a set of hard spheres that flow through a membrane, which is conceived as a collection of parallel planes with circular pores at random positions. It uses different diameters according to specified size distributions of particles and pores in the membranes. In the present work, the size distributions of both, juice particles and membrane pores are well described by a log-normal distribution (equation 1),

$$f(x) = \frac{1}{x \ln 10 \sqrt{2\pi\sigma^2}} \exp\left(-\frac{[\log(x) - \log(\mu)]^2}{2\sigma^2}\right) \quad (1)$$

The probability density function (PDF) is completely described by two parameters, μ (μm) and σ (μm), which are the mean and standard deviation of the corresponding Gaussian distribution of the common logarithm of the diameter (log (x)).

Once with the experimental data of unfiltered juice particles, membrane pores and filtered juice particle size distributions described by (μ_s, σ_s), (μ_p, σ_p), and (μ_f, σ_f), respectively, it was possible to validate the model. Thus, (μ_s, σ_s) and (μ_p, σ_p) were employed as input for the simulations in order to predict the filtered juice particle size distribution (f-SD: μ_f, σ_f) from the average of 10 independent runs each one with 1000 spherical particles. The particles were contained in a cubic box of volume (V) such that the concentration of spheres equaled the SJ or CJ concentration. The particles were made to diffuse through a membrane conformed by 5 parallel planes separated by a distance equal to the largest pore size for that system. Then, after some hundreds of thousands of simulation steps (500,000, equivalent to 5000 dimensionless time units), there were a sufficiently large number of particles at the right side of the box to consider it a statistical sample from where the filtrate particle size distribution could be obtained (Marrufo-Hernández *et al*., 2018). Finally, to validate the model; filtered juice particle size distributions (f-SD) from the simulations were compared to experimental distributions for each filtration system.

3 Results and discussion

3.1 Membranes pore size distribution (p-SD) characterization

SEM was used to characterize the cellulose acetate membranes (Zhao *et al*., 2000). Fig. 1a and Fig. 1b show the structure of M1 and M2 respectively. Both membranes show multilayers of pores of various sizes, which agree with the general conception of the membrane in the computational model.

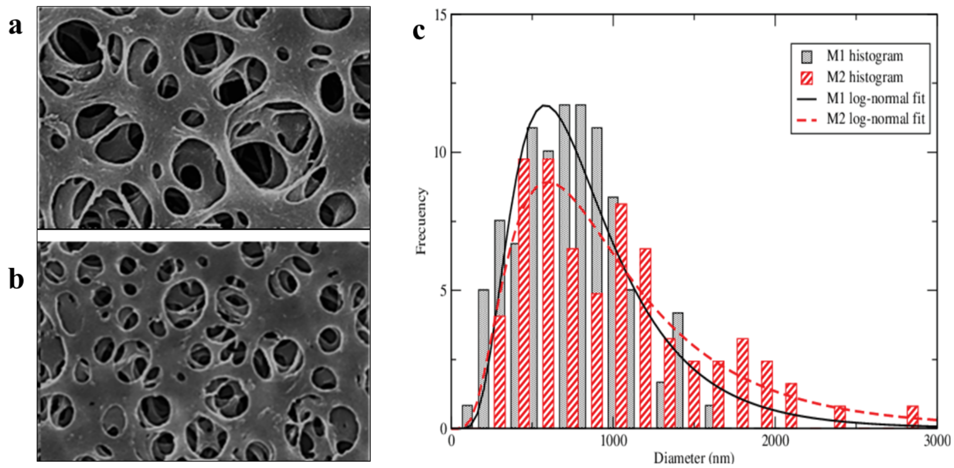

Fig. 1. Microscopy images of CA membranes: (a) M1, (b) M2. Their corresponding pore size distributions are plotted in (c): (M1 shaded bars and M2 diagonal stripes bars) showing a well-defined log-normal shape (M1 solid black line and M2 dashed red line).

The measured pore size distributions are shown in Fig. 1c as a histogram representing the frequency of pore diameters, along with the corresponding fits.

Both membranes show a very characteristic log-normal type distribution ($R^2\sim0.92$), as those reported for other CA membranes (Kellenberger *et al*., 2014). The corresponding parameters for the M1 and M2 fittings are μ_{pM1}=0.757 μm, σ_{pM1}=0.224 μm; and μ_{pM2}=0.879 μm, σ_{pM2}=0.27 μm, respectively. The corresponding mean pore diameters (D) are D_{M1}=0.865 μm and D_{M2}=1.07 μm. At first sight, size distributions of both membranes look similar, with the maximum located at the same value, and with a slight difference in polydispersity. But at this point, it is important to notice that in the case of log-normal distributions, the first momentum (mean) does not agree with the maximum, and is related to μ and σ according to equation 2

$$D = \mu 10^{\frac{\sigma^2}{2}\ln 10} \tag{2}$$

Hence, a value of σ_{pM2} being 20% larger than σ_{pM1} implies a value of D_{M2} 24% larger than D_{M1}. Furthermore, while one membrane has a mean pore size practically equal to that of the particles (see section 3.3), the other has a mean pore size considerably larger. As it is forward discussed, this selection of membranes allows to obtain valuable information concerning the role of the interactions present in the system.

3.2 Juices characterization

3.2.1 Parameters

From the juice characterization SJ showed sugar content of 15 °Bx (150 g_{sugar}/L), pH of 4.06 and RI of 1.355. On the other hand, the sugar content of the CJ batch was 11.5 °Bx (115 g_{sugar}/L), pH of 3.5 and RI of 1.350. All the results were in the range of previously reported values for apple juice (Eisele and Drake, 2005) within the accepted by Official Mexican Norms (NOM, 2009) and remained the same after the filtration process.

3.3 Juices particle size distribution prior and after filtration

The effect of pre-treatment (sedimentation and centrifugation) and filtration in the particle size distribution of apple juice samples studied by light scattering and SEM is shown in Fig. 2. Particle size distribution parameters (μ_s, σ_s) obtained from light scattering for SJ and CJ prior and after filtration are listed in Table 1. The corresponding mean particle size diameters are also included for reference.

As it can be seen from the results, the distribution of SJ (Fig. 2a) has a higher polydispersity than CJ (Fig. 2d), which means that centrifugation eliminates larger debris (in this case >2 μm) more efficiently but keeps a good content of particles as previously reported by Carbonell *et al*., 2011.

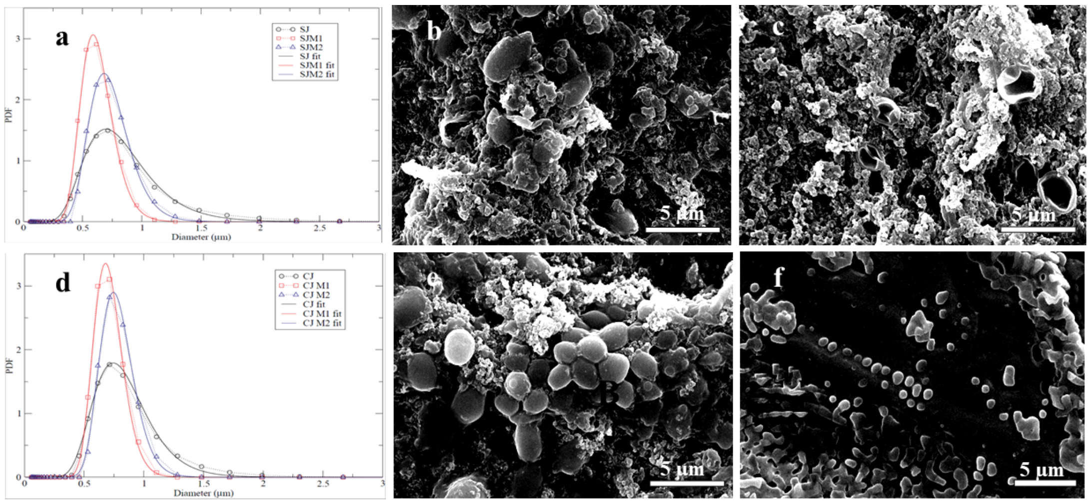

Fig. 2. Juice particle size distributions analyzed with light scattering for SJ (a), CJ (d) before (black circles) and after filtration with membranes M1 (red squares) and M2 (blue triangles) and SEM prior (b, e) and after filtration (c, f) for SJ and CJ respectively.

Table 1. Log-normal parameters and mean particle diameters of SJ and CJ apple juice obtained by light scattering prior and after filtration.

Sample	μ_f (μm)	σ_f (μm)	D (μm)
SJ	0.790	0.154	0.841
SJ M1	0.616	0.094	0.631
SJ M2	0.720	0.102	0.740
CJ	0.807	0.125	0.841
CJ M1	0.700	0.075	0.711
CJ M2	0.770	0.079	0.783

For both GD apple juices (SJ and CJ), the effect of the filtration is evident, giving as a result a narrower particle size distribution. Compared to the initial particle size distributions, the mean size of the filtered juices decreased 25% (SJ-M1), 12% (SJ-M2), 15% (CJ-M1) and 7% (CJ-M2), respectively. It is worth mentioning that all the results were in the range of previously reported particle sizes (0.47 to 0.91 μm) for some commercial apple juices (Beveridge *et al.*, 1998).

From the graphs, it can be seen that the particle size distribution of SJ juice, that was originally wider (σ_f =0.154) than that for CJ (σ_f =0.125), was consistently also wider upon filtration (σ_{fM1} =0.094 and σ_{fM2} =0.102) than that for CJ (σ_{fM1} =0.075 and σ_{fM2} =0.079). Although SJ and CJ initially had the same mean diameter, as can be observed in Fig. 2a, Fig. 2d and Table 1, the larger polydispersity of SJ the more particles were trapped by the membrane and consequently, the mean size of particles after filtration is smaller for the sedimented juice (SJ-M1 and SJ-M2) compared to their centrifuged counterparts (CJ-M1 and CJ-M2). The fact that the mean diameters of both samples are the same before filtration allows a clearer comparison of the effects of every membrane on the filtration process.

Fig. 2 also shows the microscopy images of SJ and CJ prior (b, e) and after (c, f) filtration with M1 respectively. As it can be observed, both SJ and CJ samples displayed particles with sizes near to the mean sizes obtained by light scattering. Also both samples showed aggregation before and after filtration, this type of aggregation has been previously observed and has been attributed to the interaction between particles, sugars, pectin and starch (Benítez *et al.*, 2010; Onsekizoglu *et al.*, 2010). The better efficiency of centrifugation in removing large debris as discussed above resulted in clearer and less aggregated samples for CJ and CJ-M1 as observed in Fig. 2e and Fig. 2f.

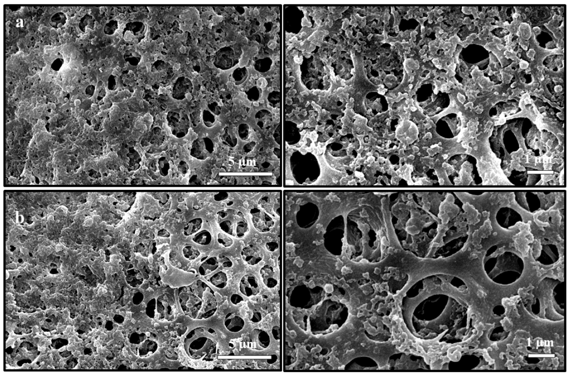

Fig. 3. SEM images of M1 after filtration of SJ (a) and CJ (b), at different magnification x5,000 and x10,000.

This behavior was expected as it has also been reported that centrifugation has a considerable effect on the reduction of turbidity (Domingues *et al*., 2014).

Finally, M1 membranes were observed after the filtration process for SJ (Fig. 3a) and CJ (Fig. 3b). Similar to the results from light scattering, M1 membranes showed a better filtration efficiency and higher reduction of polydispersity compared to M2. As it can be observed, after being used for filtration, both membranes exhibit a layer of fouling but pore size reduction is more evident as expected after the filtration of SJ compared to CJ. It has been reported that pulp content (Qiu and Rao, 1988) and average particle size (Rao *et al*., 1986) in unfiltered juices is directly related to fouling in membranes that is considered a negative factor in juice processing (de Bruijn *et al*., 2002; Domingues *et al*., 2014; Gulec *et al*., 2017) and can potentially eliminate some compounds and bigger particles than the membrane itself (Mirsaeedghazi *et al*., 2010). It can be assumed that the combination of this fouling layer and a highest pore density in the membranes with smaller pore sizes compared to the average size of juice particles (Fig. 1) allowed the reduction of particle sizes after filtration (Table 1).

3.4 *Computational simulation of apple juice filtration*

From the experimental results, it can be concluded that: i) the filtration fluid (i.e. apple juice) is conformed by quasi-spherical particles of different sizes (Fig. 2a), ii) the membrane idealization implemented in the computer simulations is an acceptable model for the CA membranes used in this work (Fig. 1); and iii) juice particle size (before and after filtration) as well as pore size distributions showed a well-defined log-normal behaviour. Under these conditions, the experimental system was considered suitable to serve as reference for comparisons with the previously reported model to evaluate its predictive capability and the validity of the assumptions made.

Experimental PDF for the aforementioned size distributions (s-SD and p-SD) have already been shown in Fig. 1 and Fig. 2 with their corresponding log-normal fittings with a coefficient of determination $R^2 > 0.99$, which means that the PDF perfectly describes the particle size distributions. Log-normal distributions have been used before in cross flow filtration modeling (Munson-McGee 2002) and membrane characterization (Kellenberger *et al*., 2014).

Fig. 4 shows the comparison between f-SD obtained from computer simulations (blue solid lines) and from experiments (red circles). As a reference, log-normal fittings for p-SD (dotted lines) and s-SD (dashed lines) employed as input for simulations are also plotted. Four filtration experiments were performed resulting from the combination of two pre-treated juice samples, SJ and CJ, being filtered with two membranes, M1 (a and c) and M2 (b and d). As mentioned before, M2 has a considerable proportion of pores larger than particles, as it can be seen in the low decaying tail of the dotted line in Figs 4b and 4d. Therefore, if only geometrical constraints were determinant in the filtration process, practically all of the particles would diffuse through the membrane. This is what simulation predicts: simulated f-SD is practically the same as s-SD, because every particle finds a pore through which it can diffuse. On the other hand, the size distribution of M1 allows to understand the effect of using a membrane with smaller pore sizes on the filtration. M1 average diameter is almost equal (barely larger) to that of juice particles. This means that the low proportion of large particles along with a low proportion of large pores leads to a very low combined probability of having one large particle finding a pore larger to diffuse on every layer of the membrane. Hence, simulations predicted an important reduction in particles larger than 1μm with the resulting reduction in f-SD polydispersity (see Figs. 2a and 2c). In both cases, according to simulations, the left side of the distributions, corresponding to smaller particles, was not affected by the filtration process, since all of these particles can diffuse through the membrane. However, the experimental f-SD shows a clear reduction in the distribution of smaller particles attributed to particle aggregation.

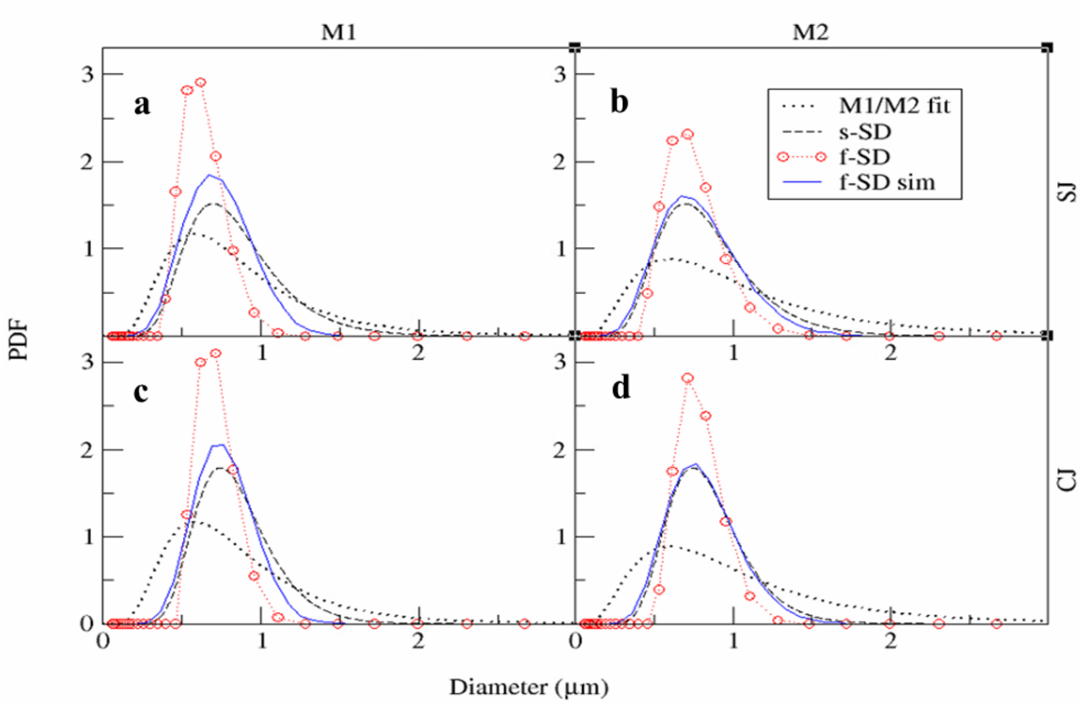

Fig. 4. Particle size distributions of the filtrate obtained from light scattering experiments (red circles) and from computer simulations (blue solid lines) for SJ and CJ, filtered with the membranes described in this work: M1 (a and c) and M2 (b and d). For reference, log-normal fittings for pore size distribution (dotted lines) and suspended solids prior to filtration (dashed lines) are plotted.

Table 2. Log-normal parameters for particle size distributions and the mean particle size of the filtrate obtained from simulations compared to experimental values.

Sample	σ_f (μm)		Error (%)	σ_f (μm)		Error (%)	D (μm)		Error (%)
	Exp	Sim		Exp	Sim		Exp	Sim	
SJ M1	0.616	0.727	18.0	0.09	0.13	40.4	0.631	0.761	20.7
SJ M2	0.720	0.767	6.5	0.1	0.15	42.2	0.740	0.811	9.6
CJ M1	0.700	0.763	9.0	0.08	0.11	48	0.711	0.788	11.0
CJ M2	0.770	0.795	3.2	0.08	0.12	54.4	0.783	0.827	5.6

On the other hand, Table 2 shows the parameters of log-normal size distributions and the mean particle size of the filtrate obtained from simulations compared to the experimental values. As it can be seen, this model has an excellent quantitative agreement in obtaining the mean particle size (D) after filtration, having a standard error around 10% or even close to 5% (CJ-M2). The only exception was for SJ-M1 with an error around 20%. On the other hand, as can be seen from the shape of PDF curves in Fig. 4 (or from the values of σ_f in Table 2) the polydispersities (σ_f) predicted by simulations are far from experimental results. When comparing the experimental and simulated PDF curves, there are two features that were observed for all systems. First, the simulation PDF curves are shifted to larger particle sizes and, second, are notoriously wider. This is what could be expected since in the computer simulation experiments, in absence of particle-particle and particle-membrane attractive interactions, the effects of a fouling layer and particle aggregation are not taken into account. Hence, the predicted filtrate will contain a higher proportion of large particles. On the contrary, in experimental systems, it is not enough for a particle to be smaller than the pore to diffuse through it. Even if a particle was smaller than a pore, still there is a probability of it being trapped in the membrane interstices either because a real membrane is not a set of planar layers, or because of chemical affinity with the membrane. This probability certainly will increase as the diameter of the particle increases. Because of that, large particles do not diffuse as could be expected considering only geometrical aspects. On the other hand, experimental distributions also seem to have a lower proportion of small particles. This may be a consequence of the aggregation of particles evidenced by SEM images. Even though this hard sphere model gives a remarkable good prediction of mean particle size and acceptable polydispersity estimations, it seems to be necessary to consider particle-particle (Israelachvili, 2009; Benítez *et al*., 2010; Genovese and Lozano, 2006) and particle-membrane (Hoek *et al*., 2003; Vrijenhoek *et al*., 2001) interactions to obtain better results. Nevertheless since many properties of interest and also membrane selection, are mainly based on mean particle size (Fitri and Widiastuti 2017; Kellenberger *et al*., 2014), this model could be a good approach for the simulation of complex systems such as apple juice filtration and could be proposed as a first attempt to provide criteria for membrane selection in the filtration of apple juice.

Conclusions

In this work, two apple juice filtration systems were studied on a lab scale and used as feed data to test and validate a novel computational simulation model. The simulations using the experimental data compared to the model gave a good estimation of the final mean particle size of the filtered juices. For the studied systems, the simulated and experimental mean pore sizes had from 79.3% up to 94.4% similarity notwithstanding the discrepancies in the polydispersity predictions due to the simplification of the interactions in the computer simulations. The obtained results suggest that this model can be proposed as a first, simple and versatile approach to provide either the characteristics of a filtrate or the characteristics of a membrane for the processing of systems of considerable complexity such as apple juice. This can represent advantages as the determinations would only require the initial characteristics of the unfiltered feed compared to a complete characterization including the feed, the membrane and the filtrate. According to the results, the model can be the base for further studies and more precise models that could include some variables such as direct particle, chemical and physical membrane-particles interactions, fouling, membrane regeneration and scaling-up to determine their application at the industrial level.

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Nomenclature

CA	Cellulose acetate
CJ	Centrifuged juice
D	Mean pore size diameter
f-SD	Filtered juice particle size distribution
GD	Golden delicious
M1	Membrane 1
M2	Membrane 2
PDF	Probability density function
p-SD	Pore size distribution
SJ	Sedimented juice
s-SD	Suspended solids size distribution
V	Volume
DLS	Dynamic light scattering
RI	Refractive index
SE	Secondary electrons

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