



Proof of concept of electrospraying skimmed milk

Bernat Perez-Playà^{a,*}, Anna Zamora^a, Llibertat Abad^b, Manuel Castillo^{a,*}

^a Centre d'Innovació, Recerca i Transferència en Tecnologia dels Aliments (CIRTA; TECNIO CERTA-UAB), Departament de Ciència Animal i dels Aliments, Facultat de Veterinària, Universitat Autònoma de Barcelona. Edifici V, Travessera dels Turons, s/n, E-08193 Cerdanyola del Vallès, Barcelona, Spain

^b Institut de Microelectrònica de Barcelona (IMB-CNM-CSIC), Carrer dels Til·lers, Campus Universitat Autònoma de Barcelona, E-08193 Cerdanyola del Vallès, Barcelona, Spain

ARTICLE INFO

Keywords:

Electrospraying
Dehydration
Skimmed milk
Non-thermal
Concentration

ABSTRACT

Electrospraying is a process that uses an electric field to remove solvents from polymeric solutions, allowing the production of micro- and nanoscale particles. This technique is characterized by its non-thermal nature, which avoids thermal damage to treated materials. This method has not been employed as a dehydration approach in food processing, and its potential applications in this field have not yet been explored or studied in depth. In this study, electrospraying for the removal of water from reconstituted milk was evaluated, yielding promising results. The increase in the distance between the tip and the collector did not show significant effects on the size of the drops due to the interaction of two opposite effects: the decrease in the intensity of the electric field and the increase in the residence time. It was also observed that a larger diameter of the tip resulted in big drops, while a high fat content generated small drops due to the increase in viscosity and decrease in surface tension. The diameter of the droplets was reduced by decreasing the flow rate and/or increasing the concentration of total solids. Finally, electrospraying, at a low flow rate, eliminated water significantly ($p < 0.05$) without producing any significant effect on the structure of proteins (i.e., caseins and whey proteins). These findings open the way for novel research directions in the drying of high-value liquid foods, highlighting the potential of this technique for innovative applications within the food processing sector.

1. Introduction

Raw milk is a fresh material very susceptible to being altered by microorganisms because it has an excellent composition of nutrients and elevated water activity, having a shelf life of 12 to 24 h (Lara-Aguilar & Alcaine, 2019). For dairy matrices, water extraction is widely recognized as a highly effective preservation method, owing to its ability to minimize microbial activity, slow enzymatic degradation, and enhance overall product stability (Mari et al., 2024). In the case of milk powder, the shelf life is extended to six months and three years, for whole and skimmed milk, respectively, while maintaining its nutritional value (Whetstone & Drake, 2007). Additionally, it has added value, as a great advantage is obtained at the logistical level, because the extraction of water leads to a much lower storage volume, reducing both stockage and transportation costs (Bocci & Casas, 2013).

Currently, industrial milk powder production is mainly based on pre-concentration by vacuum evaporation followed by spray drying, due to its high throughput, continuous operation, short residence times, and proven scalability. However, this route is energy-intensive and involves

thermal exposure, which may affect heat-sensitive milk constituents such as whey proteins, enzymes, vitamins, lactose, and mineral equilibria depending on processing parameters (Fox et al., 2015; Patil et al., 2021). Low-temperature alternatives such as membrane concentration can reduce the water content before drying and better preserve native protein functionality, although they are limited by fouling, concentration polarization, increasing viscosity, and restricted final solids concentration (Deshwal et al., 2021; Kumar et al., 2013). Freeze-drying provides high-quality products and good preservation of heat-sensitive compounds, but it is constrained by long processing times, high energy demand, batch operation, and low throughput (Ratti, 2001). Roller drying is robust and scalable, but direct contact with heated surfaces may intensify thermal damage and reduce powder functionality (Carić et al., 2009). In this context, electrospraying may offer a non-thermal alternative for water removal from liquid foods, with potential advantages for preserving protein functionality, although its processing capacity, energy efficiency, scalability, and ability to produce fully dried dairy powders remain to be demonstrated (Bhushani & Anandharamakrishnan, 2014; Dhiman et al., 2022).

* Corresponding authors.

E-mail addresses: Bernat.perez@uab.cat (B. Perez-Playà), Manuel.Castillo@uab.cat (M. Castillo).

<https://doi.org/10.1016/j.ifsset.2026.104757>

Received 1 May 2026; Received in revised form 30 July 2026; Accepted 24 August 2026

Available online 24 August 2026

1466-8564/© 2026 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY-NC license (<http://creativecommons.org/licenses/by-nc/4.0/>).

Electrohydrodynamic atomization (also called electrospraying) is a technique that generates, under the influence of an electrical field, fine droplets with a monodispersed size distribution from a liquid (Barrero & Loscertales, 2007). In this technique, the energy of an intense electric field, in the order of $\text{kV}\cdot\text{m}^{-1}$, is used to achieve the separation of microdroplets. Once charged droplets have been formed, they undergo a shrinkage process due to solvent evaporation (Bohr et al., 2013). When this electrical concentration of the charged droplet reaches the Rayleigh limit, the balance between surface tension and electrical forces is broken, and Coulomb fission occurs causing the droplet to fragment into smaller droplets and partial solvent vaporization (Zhou & Biswas, 2021). There are several parameters that affect the performance of the electrospraying process: solution parameters and processing parameters. The solution parameters that influence the process include surface tension, density, viscosity, and electrical conductivity. Regarding, processing conditions, the applied voltage and the distance between the tip and the collector affects the electric field architecture (Bohr et al., 2013).

Electrosprayed materials are widely studied in the fields of tissue engineering and regenerative medicine, packaging, nanostructured sensors, textile industry, materials science, energy materials, films technologies, and chemistry (Landa, 2017). Moreover, the electrosprayed particles have functional advantages due to their microstructure obtained through exposure to an electric field (Bhushani & Anandharamakrishnan, 2014; Xie et al., 2014). The use of electrospraying process in the field of food processing and preservation remains largely unexplored (Ghorbani & Santos, 2021). In food processing, electrospraying may be of great interest since it avoids the use of extreme temperatures (hot and cold -i.e., in freeze drying-); it significantly reduces energy consumption compared to prevailing methods in the industry for heat-sensitive products, all while preserving the food's nutritional and functional qualities, in line with stringent food safety standards (Persano et al., 2013).

The present study aimed at evaluating the application of electrospraying as a potential alternative method for water removal from milk, as a proof of concept. The principal objective was further subdivided into three specific objectives to ensure a comprehensive and detailed examination of the process. Firstly, the influence of key processing parameters in the electrospraying method was evaluated, including the effect of distance between tip-collector and the effect of milk fat, among others. Secondly, the degree of water extraction in the skimmed milk processing by electrospraying was determined. Thirdly, the effect of the electrospraying process on main milk proteins, both caseins and whey proteins, was analyzed.

2. Material and methods

2.1. Electrospraying equipment setup

The equipment used in this work was the Fluidnatek LE-10 (Bio-inicia, Valencia, Spain), which has a horizontal configuration between

tip and collector. Fig. 1 shows the equipment configuration and the different components that constitute the system. This equipment was located in the clean room (*Instalación científico técnica singular-ICTS-CSIC*) of the IMB-CNM-CSIC, where there is controlled atmosphere (positive pressure with respect to atmospheric pressure, relative humidity at $40 \pm 5\%$ and the temperature at $20 \pm 2^\circ\text{C}$). The equipment was modified for this experimental study to incorporate the NE-4000 syringe pump (New Era Pump Systems Inc., Farmingdale, United States).

2.2. Preliminary tests

Three preliminary tests were conducted for exploratory purposes.

Firstly, a screening test was performed to monitor the effect of flow rate (0.005 to $0.2\text{ mL}\cdot\text{min}^{-1}$) on the droplet size, with an applied potential of 29 kV and a distance tip-collector of 10 cm using skimmed milk powder (LACTALIS Ingredients, Bourgbarré, France) reconstituted with ultra-pure water (Mili-Q water with a resistivity of $18\text{ M}\Omega\cdot\text{cm}$) at 12% of total solids (see Section 2.4).

Secondly, the impact of the fat content in the injected milk was evaluated to determine whether it was triggering difficulties due to electrical insulator capacity it has. Pasteurized skimmed, semi-skimmed and whole commercial milk (Hacendado, Naturleite, Lugo, Spain) was used and different conditions were evaluated, including flow rate (0.01 and $0.075\text{ mL}\cdot\text{min}^{-1}$) and outer tip diameter (1.59 and 3.18 mm).

Thirdly, it is well known that the distance between tip and collector is also a critical point on the electrospraying process in material science. The intensity of electrical field decreases with the inverse of the square of this distance, but on the other hand, the rise of distance induces an increment of residence time what would cause a greater dehydration. The effect of distance tip-collector (10 to 22 cm) on skimmed milk processing was evaluated using reconstituted skimmed milk at 12% of total solids with and applied potential of 29 kV and a flow rate of 0.01 and $0.075\text{ mL}\cdot\text{min}^{-1}$.

During each trial, the electrospraying regime was visually monitored from the droplet formation behavior at the nozzle tip. The operating mode was assessed qualitatively based on the observed shape and stability of the meniscus/Taylor cone and the droplet ejection pattern.

2.3. Experimental design

Based on the preliminary tests, two flow rates that showed clearly different process responses were selected for the final experimental design: 0.01 and $0.2\text{ mL}\cdot\text{min}^{-1}$. A non-randomized factorial experimental design was applied to this work, comprising three skimmed milk reconstitution levels (12 , 24 and 30%) and the two selected flow rates (0.01 and $0.2\text{ mL}\cdot\text{min}^{-1}$) to evaluate the effects of these on protein denaturation and total solids concentration (Fig. 2). The voltage (29 kV) and the distance between the tip and the collector (10 cm) were the same throughout the study. The experiment was replicated twice, resulting in a total of 12 trials.

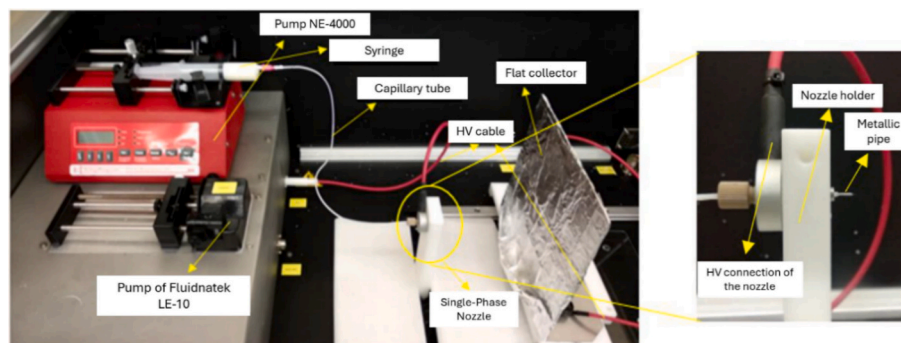


Fig. 1. Setup view of the final configuration and components of the Fluidnatek LE-10 system equipped with NE-4000 syringe pump.

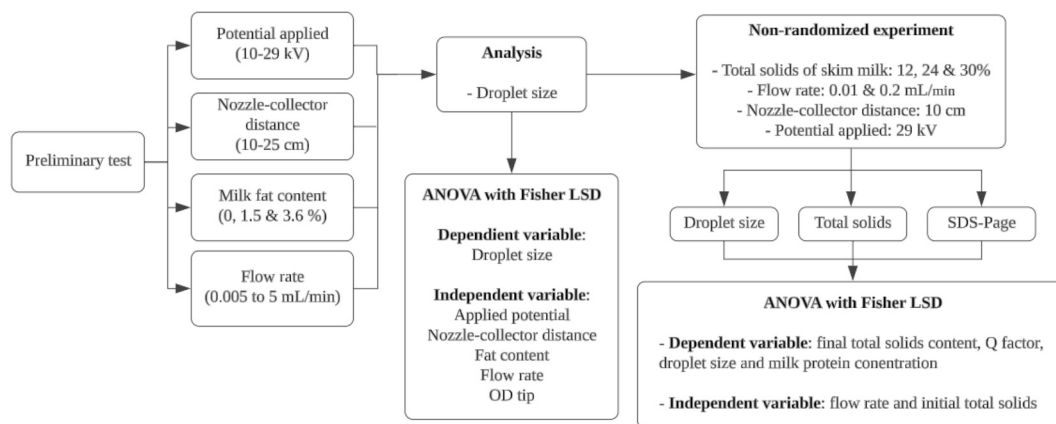


Fig. 2. Diagram of the implemented experimental design.

A nozzle–collector distance of 10 cm was selected for the final experimental design because it provided more reliable deposition on the collector under the tested conditions. Larger distances did not significantly reduce the deposited droplet size and progressively worsened the deposition pattern, with partial product loss observed from 20 cm onwards.

2.4. Skimmed milk powder reconstitution

A commercial skimmed milk powder (LACTALIS Ingredients, Bourgarré, France) dehydrated by spray drying was used. It is a standard milk with a high microbiological and functional quality and a Whey Protein Nitrogen Index (WPNI) $\geq 7 \text{ mg} \cdot \text{g}^{-1}$. The reconstitution of the milk was carried out individually with Milli-Q water (resistivity: 18 M Ω ·cm) at 20 °C and the amount of added milk powder was adjusted to reconstitute it at 12, 24 and 30 (w/w). Weighing of the milk powder and the water was done with an analytical balance (FV-220C, Gram Group, Barcelona, Spain) with a precision of 0.1 mg. The reconstitution process was based on three stages: 30 min of stirring, 30 min undisturbed under darkness, and 5 min of sonication to eliminate small air bubbles that could affect the correct operation of the electrospraying process. The beaker where the milk was reconstituted was kept closed, to avoid the evaporation of water due to the difference in relative humidity between the sample and the environment.

2.5. Deposited product droplet size

A visible light reflection optical microscope (Leica DM2700M, Leica Microsystems, Wetzlar, Germany) in transmission operation was used to carry out the droplet size analysis. It has a built-in video camera (Moticam 10+ SB-09, Motic, Hong Kong, China). Images, with a

resolution of 1832 $\times$ 1372 pixels, were taken using a 5 $\times$ objective lens, resulting in a total magnification of 20 $\times$ in order to observe the droplets. The image software used was Motic Image plus 3.0. Glass microscope slides of 20 $\times$ 20 $\times$ 1 mm were used and covered with glass coverslips prior to observation.

For each experimental trial, two glass slides were prepared. Five images were acquired from each slide at different positions: upper, central, lower, left, and right areas. Therefore, a total of ten images were analyzed per experimental trial. Droplet size analysis was performed using ImageJ software (version 1.54 g, <https://imagej.net/ij/>). The diameter of individual droplets was measured manually from calibrated images using the line measurement tool (Fig. 3). Droplets that were out of focus, overlapping, irregularly spread, or partially located at the image borders were excluded from the analysis. The average droplet diameter for each trial was calculated from the individual droplet measurements obtained from the ten images.

2.6. Quantification of water removal achieved by the electrospraying process

The water removal degree was evaluated using the Q factor, which is a dimensionless parameter, defined by the ratio between the dry matter content of the concentrated product and that in the original material (Walstra et al., 2006a). Q factor was calculated from initial and final total solids (TS_i and TS_f , %, respectively; Eq. (1)).

$$Q \text{ factor} = \frac{TS_f}{TS_i} \quad (1)$$

The initial total solids were calculated in triplicate by measuring approximately 1 g of product that was placed into an aluminum capsule, using an analytical balance FV-220C. The product was completely dried

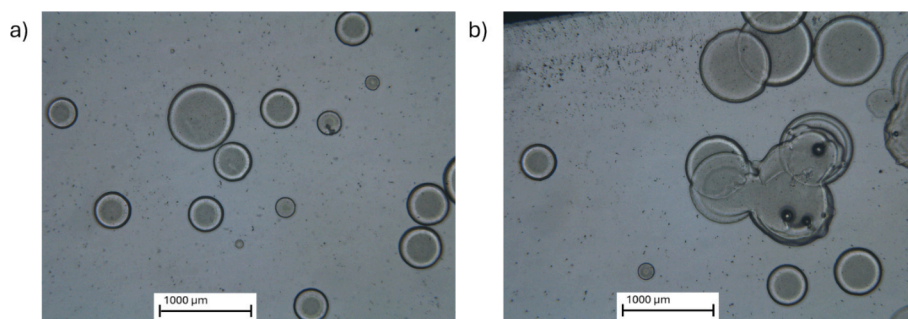


Fig. 3. Optical microscopy images with magnification factor of 20 $\times$. Skimmed milk reconstituted at 12% electrosprayed at 29 kV and a flow rate of 0.01 mL·min $^{-1}$ and 10 cm of nozzle–collector distance; a) Field showing isolated deposited droplets included in the manual ImageJ diameter analysis; b) field showing overlapping, coalesced, or irregularly spread droplets that were excluded from the analysis.

using a convection oven at 105 °C (UFB 400, Memmerk GmbH, Schwabach, Germany) for 24 h to reach constant weight. The total solids content (TS, %) was calculated as the ratio between the weight of the sample after complete dehydration (w_f , g) and the initial weight of the sample before drying (w_0 , g) as:

$$TS = \frac{w_f}{w_0} \cdot 100 \quad (2)$$

The final content of total solids was determined using the procedure described in the previous section with the difference that the sample was taken, weighed and dried directly with a weighed aluminum foil previously dehydrated for 2 h at 105 °C in a convection oven (Fig. 1; aluminum foil covering the flat collector). Since different injection electrospraying flows were studied, the time required to take 1–4 g of final product was adjusted to 20 and 180 min, for the flow rates 0.2 and 0.01 mL·min⁻¹, respectively. The total solids content of the sample was calculated as previously described by Eq. (2). Subsequently the Q factor index was determined.

2.7. Evaluation of the effect of electrospraying on milk proteins

Both qualitative and quantitative analysis of the main proteins of the electrosprayed samples were carried out by SDS-PAGE under reducing and non-reducing conditions using separating and stacking gels containing 14% or 4% acrylamide, respectively. The reducing conditions were achieved by the addition of a reducing reagent, β-mercaptoethanol, that breaks disulfide bridges (Laemmli, 1970). Reduced and non-reduced samples were mixed twice their volume with double-strength buffer containing 0.5 M Tris, 2% sodium dodecyl sulphate (SDS), 10% glycerol and 0.025% bromophenol blue. For reducing conditions, a 5% β-mercaptoethanol was added on buffer solution. The prepared samples were incubated 5 min at 45 and 95 °C, for non-reducing and reducing conditions, respectively, and centrifuged at 2500 xg for 30 min (Freire, 2022). Supernatants containing the protein without fat were diluted with the same buffer to have a dilution 10⁻¹ and 15 μL was loaded onto the gels.

Bovine serum albumin (BSA, 10 μg) and a mix of casein (CN, 10 μg), α-lactalbumin (α-LA, 10 μg) and β-lactoglobulin (β-LG, 10 μg) were used as standards and were studied together with the samples. Gels were run at 200 V, stained using 0.1% (w/v) Coomassie Brilliant Blue R-250 in a 16:1:50 mixtures of methanol, acetic acid and distilled water. Distained gels were scanned with CanonScan (Lide 220, Canon, Japan) and analyzed with GelAnalyzer software (version 19.1, www.GelAnalyzer.com). The amount of sample (15 μL) was determined with trial-error test to obtain bands large enough for the whey proteins without reaching the total saturation of the casein bands, because the percentage of caseins is much higher than that of whey proteins.

2.8. Experimental results statistical analysis

Experimental data were analyzed using the OriginPro 2018 software (version b9.5.1.195, OriginLab, Northampton, Massachusetts, United States).

The evaluation of the effect of selected process variables (independent variables) on key response variables (dependent variables) was performed using ANOVA, followed by Fisher's LSD mean comparison test to identify significant differences ($p < 0.05$) between treatment levels. In all cases, ANOVA was performed considering only the experimental factors to avoid overparametrisation. The experimental sequence was non-randomized due to operational constraints related to sample preparation, equipment stabilization, and the long processing times required at low flow rate. To assess the potential influence of run-to-run variability, replicate was initially included as a blocking term in preliminary ANOVA models. Since no significant replicate effect was observed, this term was not retained in the final models.

The linear models used were $Y_i = \mu + F_i + \varepsilon_i$ for the one-factor ana-

lyses and $Y_{ij} = \mu + A_i + B_j + (AB)_{ij} + \varepsilon_{ij}$ for the two-factor analyses. In the one-factor models, droplet size was the dependent variable and nozzle-collector distance, fat content, flow rate, or OD tip was included separately as the independent variable. In the two-factor models, droplet size and final total solids were the dependent variables, and initial total solids and flow rate were the independent variables. Linear fittings shown in the preliminary tests were used only as descriptive indicators of experimental trends and were not intended as predictive models. The effects of the experimental factors were considered significant at $p < 0.05$, and mean comparisons among treatment levels were performed using Fisher's LSD test.

3. Results

3.1. Preliminary test

3.1.1. Effect of flow rate of the electrosprayed solution on droplet size

A significant, positive correlation between flow rate and droplet size ($p < 0.05$) was observed when the applied potential during the electrospray process was 29 kV with a tip-collector distance of 10 cm in the processing of reconstituted skimmed milk at 12% (w/w) (Fig. 4). The initial droplet diameter at the breakup position increases proportionally to the cube root of the flow rate, explaining the observed experimental trend (Hartman et al., 2000). At higher flow rates, a greater liquid volume reaches the tip per unit time, which decreases the total electric charge per droplet. Consequently, the droplets underwent less effective Coulomb fission, resulting in a larger average size upon reaching the collector (Rayleigh, 1882; Zhou & Biswas, 2021). Furthermore, the obtained data showed polydispersity, as observed by optical microscopy. Previous studies have shown that when the electrospray conditions were kept constant, the particle size was monodispersed, finding most particles in a certain range (Bhushani & Anandharamakrishnan, 2014; Hernández, 2018). In this study, droplets rather than particles were obtained due to a non-optimized electrospraying process and non-optimized environmental conditions. Still, during the splitting processes, droplets broke up into smaller droplets and there were no significant differences in mean droplet size due to small changes in the flow rate, e.g. from 0.005 to 0.01 mL·min⁻¹, which is consistent with previous results.

The magnitude of the droplet size changes observed in the present study should be interpreted with caution, since the measured structures were deposited wet or partially concentrated droplets rather than fully dried electrosprayed particles. In many electrospraying studies, size is

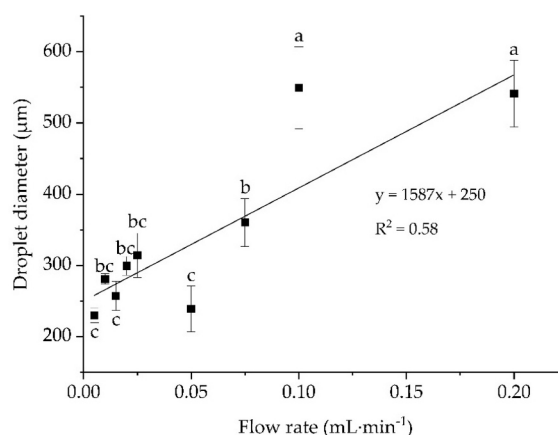


Fig. 4. Relationship between average droplet size and flow rate when processing skimmed milk reconstituted at 12% with an applied potential of 29 kV and a distance tip-collector of 10 cm. Number of replicates, $n = 3$; Number of observations, $N = 27$; Mean value $\pm$ s.e values without common superscripts were significantly different ($p < 0.05$).

reported for particles after solvent evaporation and solidification under stable cone-jet conditions, where flow-rate effects are commonly described through scaling laws for initial jet breakup and droplet formation (Hartman et al., 2000), followed by drying, charge redistribution, and Coulomb fission processes (Bohr et al., 2013; Zhou & Biswas, 2021). In contrast, in the present work, skimmed milk was processed as a complex aqueous food matrix and the collected material retained residual moisture. Therefore, the measured diameter may reflect not only primary atomization but also differences in residence time, evaporation extent, charge-to-mass ratio, repeated Coulomb fission events, droplet spreading on the collector, and possible coalescence during deposition (Rayleigh, 1882; Zhou & Biswas, 2021). These factors may amplify the apparent effect of flow rate on the final deposited droplet diameter, making direct comparison with studies on dried electro-sprayed particles inappropriate.

3.1.2. Effect of fat concentration and tip diameter on electro-spraying process and droplet size

As expected, in processing tests with different fat concentrations, it was observed that an increase in the tip diameter from 1.59 to 3.18 mm led to a significant ($p < 0.05$) increase in the average diameter of the fat drops (Fig. 5a) since the initial drop formed in the tip was larger, which caused the resulting drops to be proportionally larger after fragmentation, as observed in the study of Jafari-Nodoushan et al. (2015), where the particle size was larger as the diameter of the tip increased. In addition, it was observed that the highest fat content (3.5%) generated significant smaller droplets but there were not significant differences ($p > 0.05$) in the small variations of fat content (0.15 and 1.5%) (Fig. 5b). This effect may be attributable not only to the slight increase in viscosity of the liquid (from 1.68 to 1.93 mPa·s for skimmed and whole milk, respectively), which tends to improve the internal droplet cohesion and also to change the interfacial properties induced by fat, such as surface tension and conductivity, that could influence droplet size (Tro, 2022; Walstra et al., 2006b).

The increase in fat content also reduced the surface tension, which varied between 49 and 60 mN·m⁻¹ in skimmed milk and 44–52 mN·m⁻¹ in whole milk (Mukherjee et al., 2005). This decrease in surface tension favors the stabilization of the Taylor cone and facilitates the fragmentation of the drops, since it reduces the Rayleigh limit, promoting greater atomization of the liquid (Zhou & Biswas, 2021). During the processing of whole milk, it could be observed that a quasi-stabilization of Taylor cone (Fig. 6a) in comparison to skimmed milk (Fig. 6b) which was more like spindle mode. Stabilization of the Taylor's cone was attributable to low surface tension of the whole milk due to the fat content (Smith, 2015). The milk inlet temperature (~22 °C) could not be increased to

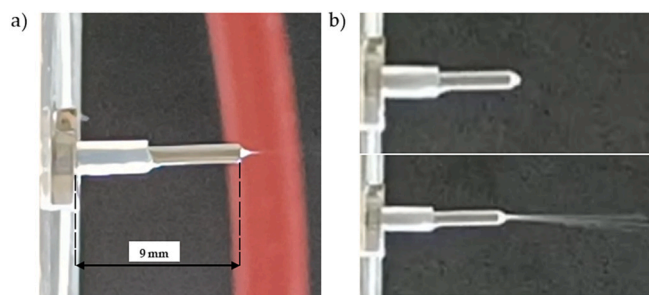


Fig. 6. a) Whole milk in quasi-stabilized cone-jet mode; b) skimmed milk in spindle mode: the top image showed the droplet formation, while the bottom image represents the subsequent droplet ejection.

~44 °C to fully liquefy the fat and mitigate potential clogging in the electro-spray process because the equipment lacked a built-in temperature control system. Therefore, part of the observed differences in deposited droplet size may be related not only to the nominal processing parameters, such as flow rate, nozzle diameter, or nozzle-collector distance, but also to changes in the spray regime. Since spray mode was assessed qualitatively and not by high-speed imaging, these mechanistic interpretations should be considered exploratory.

3.1.3. Effect of distance on electro-spraying process and droplet size

It was hypothesized that an increase in the distance between the tip and the collector would result in a decrease in droplet size due to the higher residence time, which would allow for greater solvent evaporation and greater droplet fragmentation (Jafari-Nodoushan et al., 2015). However, the experimental results showed no significant correlation (F value ~ 1.09 and p value ~ 0.40) between distance and droplet diameter (Fig. 7). This phenomenon could be explained by the reduction in the intensity of the electric field as the distance increases, if the applied potential and the flow rate are kept constant. At greater distances, the lower intensity of the field reduces the disruption force on the droplets, which tends to reduce the number of fragmentation processes (Jiang & Yu, 2018). These effects (i.e., decrease of intensity and increase of residence time with distance) could counteract each other, explaining the absence of significant variations in the size of the droplets observed in the experiment. Another effect of water evaporation must be considered, which reduces the size of the drop, causing a greater charge per unit of surface, favoring the electrical instability of the drops, giving rise to a greater number of fissions (Bohr et al., 2013).

During the experiment, it was observed that as the distance between

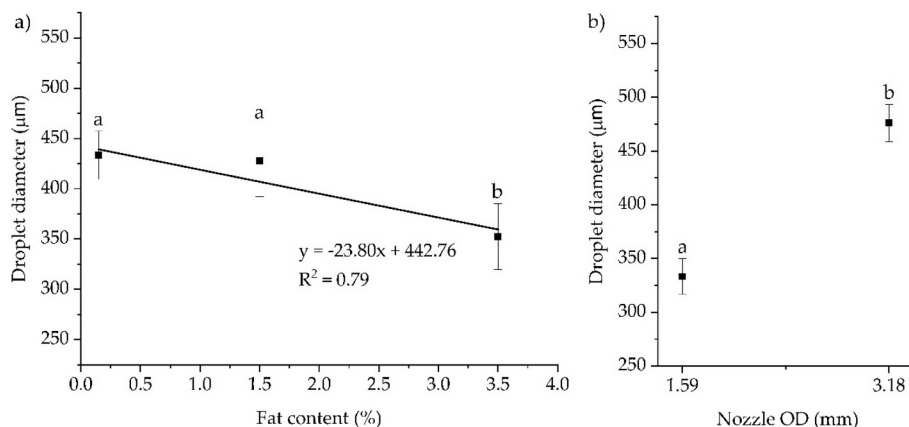


Fig. 5. Relationship between droplet size and milk fat content after electro-spraying process: a) droplet size vs fat content; and b) droplet size vs tip diameter. Number of replicates, $n = 4$; Number of observations, $N = 24$; a) Least squares means for each fat level is based on average of 8 trials over a range of 2 OD levels; b) Least squares means for each OD is based on average of 12 trials over a range of 3 fat levels. LMS value $\pm$ s.e.; a-b values per fat content or tip OD without common superscripts were significantly different ($p < 0.05$).

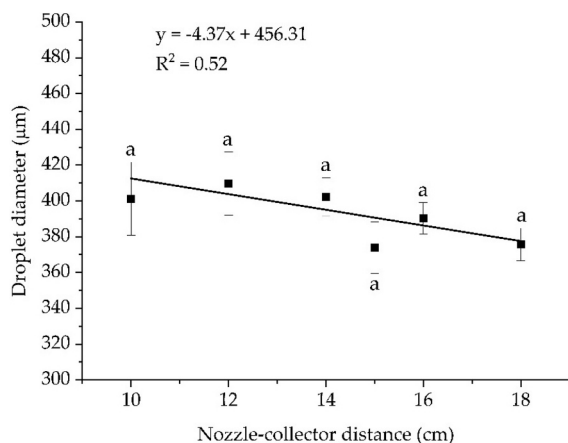


Fig. 7. Relationship between droplet size and distance tip-collector on the electrospraying process using reconstituted milk at 12%. Number of replicates, $n = 2$; Number of observations, $N = 24$; Mean value $\pm$ s.e.; a-b values without common superscripts were significantly different ($p < 0.05$).

the tip and the collector in the horizontal electrospraying configuration was increased, a progressive change in the deposition pattern of the product occurred (Fig. 8a). Specifically, a greater proportion of the material tended to accumulate at the bottom of the collector as the distance was increased. From 20 cm onwards, part of the product was no longer able to reach the collector and fall to the base of the equipment (Fig. 8b).

3.2. Final product characterization

3.2.1. Degree of water removal

Electrospraying was able to remove water from milk, and the degree of water removal depended on both flow rate and initial total solids (Table 1). At $0.2 \text{ mL} \cdot \text{min}^{-1}$, the final total solids were similar to those of the initial milk, resulting in Q factors close to 1 regardless of the initial concentration. In contrast, reducing the flow rate to $0.01 \text{ mL} \cdot \text{min}^{-1}$ led to significant water removal ($p < 0.05$), with Q factors of approximately 3, 2, and 2 for milk reconstituted at 12, 24, and 30%, respectively. The highest relative concentration was therefore obtained for milk reconstituted at 12% and processed at $0.01 \text{ mL} \cdot \text{min}^{-1}$. This behavior can be explained by the coupling between electric field intensity, charge accumulation, droplet breakup, and evaporation. At lower flow rate, the same applied voltage acts on a smaller liquid volume per unit time, increasing the effective charge-to-mass ratio and the surface charge density of the emitted droplets (Rebollo, 2015). This enhances electrostatic stresses at the droplet surface, favoring deformation, breakup, and solvent evaporation when these stresses overcome the stabilizing effect of surface tension (Gomez & Deng, 2011; Xie et al., 2014). As evaporation proceeds, the droplet radius decreases and the relative surface charge density increases, which may promote further instability and Coulomb fission when the Rayleigh limit is reached (Bohr et al., 2013; Rayleigh, 1882). Therefore, the higher Q factor at $0.01 \text{ mL} \cdot \text{min}^{-1}$ can be

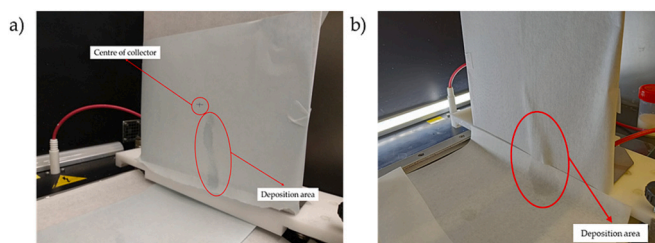


Fig. 8. Deposition of skimmed milk, when the distance tip-collector was a) 20 cm; and b) 22 cm.

Table 1

Total solids, Q factor and droplet diameter of milk samples reconstituted at 12, 24, and 30% and electrosprayed at two different flow rates.^{a-f}

Flow Rate ($\text{mL} \cdot \text{min}^{-1}$)	Reconstituted milk (%)	Electrosprayed milk		
		Total solids (g TS/100 g milk)	Q factor [†] (dimensionless)	Droplet diameter (μm)
0.2	12%	12.32 ± 0.06^f	1.055 ± 0.007^c	377 ± 74^a
	24%	23.55 ± 0.78^e	1.015 ± 0.035^c	269 ± 64^{ab}
	30%	28.94 ± 0.30^d	0.995 ± 0.007^c	156 ± 155^b
0.01	12%	36.77 ± 4.62^c	3.160 ± 0.396^a	283 ± 161^{ab}
	24%	48.54 ± 2.96^b	2.080 ± 0.127^b	169 ± 60^b
	30%	61.88 ± 1.93^a	2.125 ± 0.064^b	148 ± 39^b

Number of replicates, $n = 3$; Number of observations, $N = 18$; Mean value $\pm$ s.d.

^{a-f} values per column without common superscripts were significantly different ($p < 0.05$).

[†] Ratio between the dry matter content of the concentrated product and that in the original milk (Walstra et al., 2006a).

attributed to a higher charge-to-mass ratio, more effective droplet fragmentation, and greater evaporative surface area. Conversely, at $0.2 \text{ mL} \cdot \text{min}^{-1}$, the higher liquid throughput likely reduced the effective charge per unit mass, limiting droplet breakup and water evaporation.

The effect of initial total solids can be interpreted through the physicochemical changes induced by milk concentration. Surface tension is one of the key parameters controlling droplet formation and solvent removal during electrospraying (Xie et al., 2014), although milk surface tension has been reported to change only slightly with solids concentration at temperatures below 60°C (Williams et al., 2005). In contrast, increasing total solids may decrease water vapor pressure and water availability, thereby reducing the relative extent of evaporation. This could explain why the Q factor was lower for milk reconstituted at 24 and 30% than for milk reconstituted at 12%, despite the higher final total solids reached at higher initial concentrations. At the same time, increasing solids concentration may increase electrical conductivity, which favors charge transport and the formation of smaller droplets during electrospraying (Hartman et al., 1999). This is consistent with the observed decrease in droplet diameter as initial total solids increased. However, higher solids concentration may also increase viscosity and modify surface tension, which can affect jet stability and droplet breakup (Hernández, 2018). Therefore, the observed trends likely result from the combined effects of conductivity, viscosity, surface tension, vapor pressure, and charge-to-mass ratio rather than from a single controlling factor. It should be noted that these conditions should not be interpreted as fully optimized operating conditions, since the present work was designed as a proof-of-concept study. Moreover, a stable cone-jet mode was not consistently achieved for skimmed milk; the process mainly exhibited spindle droplet ejection. The observed dehydration cannot be directly attributed to stable Taylor cone formation. In electrospraying, the operating regime depends on the balance between electrostatic forces, surface tension, viscosity, conductivity, and flow rate, which collectively determine jet breakup, droplet charge, and droplet size distribution (Barrero & Loscertales, 2007; Hartman et al., 1999, 2000). The improved water removal observed at low flow rate was more likely related to a higher charge-to-mass ratio and greater evaporation from smaller charged droplets, according to the Rayleigh instability and Coulomb fission mechanisms (Bohr et al., 2013; Rayleigh, 1882; Zhou & Biswas, 2021).

3.2.2. Effect of electrospraying on milk proteins

The effect of electrospraying in proteins was evaluated with two SDS-PAGE gels under reducing and non-reducing conditions (Fig. 9). The same bands were obtained for all the samples, and the only variation corresponded to their intensity and thickness, directly related to the protein concentration in each sample. This occurred because the protein concentration varies among the samples and is directly associated with

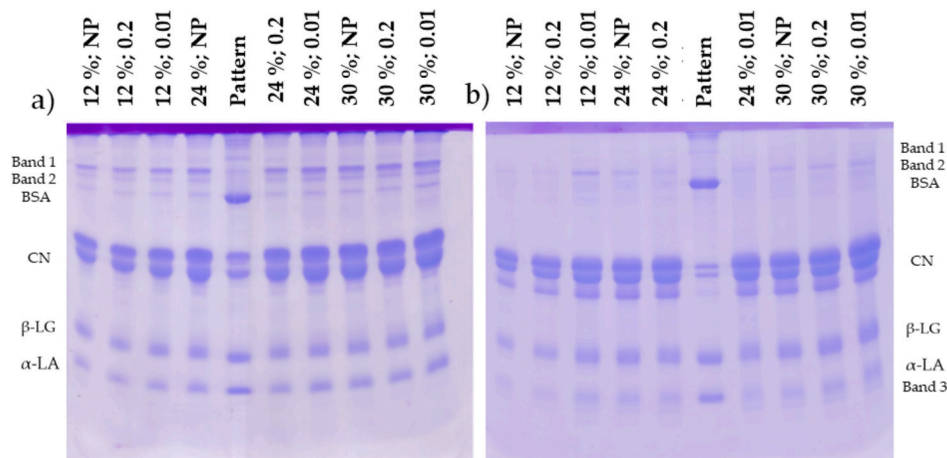


Fig. 9. SDS-Page profiles of milk reconstituted at 12, 24 and 30% and flow rates of 0.2 and 0.01 mL·min⁻¹. a) Non-reducing conditions; b) Reducing conditions. NP: non-processed; BSA: Bovine serum albumin; CN: casein; β-LG: β-lactoglobulin; α-LA: α-lactalbumin.

the extent of water removal. As expected, the intensity and thickness of the bands increased as the percentage of solids increased in milk reconstituted at 12, 24 and 30%.

In between the stacking and running non-reducing gels, some material became obstructed for all samples (Fig. 9a); these proteins or aggregates had a molecular weight too high to run on the gel, and for this reason they remained anchored. They could be associated with proteins of the membrane of the fat globule or with proteins linked together by disulfide bridges, as they mostly disappear in the gel under reduced conditions (Fig. 9b). Band 3, which only appears under reducing conditions and shows a molecular weight lower than α-LA could be associated with gamma caseins, degradation products of caseins (Eskin & Goff, 2013). During thermal treatments designed to remove water from milk, where protein denaturation occurs, at a fundamental level, protein denaturation is often defined as any noncovalent change to the secondary or tertiary structure of the protein molecule. Subsequently, whey proteins do not appear in their proper place on the polyacrylamide gel because they form aggregates between them through disulfide bridges (Schuck, 2009). For this reason, they remain in the top part of the gel in non-reduced conditions; and when reducing them with β-mercaptoethanol, new bands appear or the intensity increases significantly, which correspond to denatured whey proteins (Jovanovic et al., 2007).

In order to evaluate the effect of electrospraying on the main milk proteins, their proportion was compared with that of the initial milk (Table 2). There were no significant differences ($p > 0.05$) between the final product and initial milk under either reducing and non-reducing conditions. So, it can be established that electrospraying did not alter the secondary or tertiary structure of milk proteins. These findings are consistent with previous studies in which electrospraying showed no detrimental effect on protein structure or activity, including work with BSA and lysozyme (Xie & Wang, 2007), lysozyme (Moreira et al., 2021),

xylanases (Zhang et al., 2024), and bovine lactoferrin (Man et al., 2022), among others. However, it is important to note that none of these previous studies involved proteins within a complex liquid food matrix, highlighting the significance of the present results for proteins in such environments.

Significant differences ($p < 0.05$) appeared only for β-LG between the non-reducing and reducing conditions. This might be due to the heat treatment used to obtain the starting milk powder during industrial processing, despite its “low-heat” grade. The most relevant finding of the protein analysis was that electrospraying did not induce additional detectable changes in the SDS-PAGE profile of the main milk proteins compared with the non-electrosprayed reconstituted milk used as the control. This result is particularly relevant because the starting material was a low-heat skimmed milk powder with WPNI >7, indicating that whey proteins were largely retained in an undenatured state. However, these results should not be extrapolated to all heat-sensitive bioactive compounds or targeted nutrients, since these compounds were not specifically analyzed in the present study.

4. Future work

Future work should focus on process optimization to increase the degree of dehydration, reduce droplet size variability, and improve overall process efficiency. Relevant strategies include optimizing flow rate, applied voltage, and nozzle–collector distance, while also controlling air temperature, relative humidity, and collector design. Particular attention should be given to stabilizing the Taylor cone or cone-jet regime in skimmed milk, since a more stable electrospraying mode may enhance droplet breakup, increase evaporative surface area, and improve dehydration efficiency. Scale-up should also be specifically addressed through multi-nozzle electrospraying configurations, continuous collector systems, or assisted drying approaches, as the single-

Table 2
Percentages of main proteins for electrosprayed product and initial milk.^{a-d}

	BSA (%)	Casein (%)	β-Lactoglobulin (%)	α-Lactalbumin (%)
Non-reducing conditions				
Final product	1.13 ± 0.35 ^a	73.25 ± 1.41 ^a	9.01 ± 0.39 ^b	6.26 ± 0.16 ^a
Initial milk	1.16 ± 0.32 ^a	74.47 ± 1.44 ^a	9.77 ± 0.11 ^b	6.41 ± 0.16 ^a
Reducing conditions				
Final product	1.04 ± 0.01 ^a	71.23 ± 2.15 ^a	14.98 ± 1.35 ^a	5.91 ± 0.59 ^a
Initial milk	1.10 ± 0.19 ^a	71.45 ± 2.68 ^a	15.46 ± 1.56 ^a	5.85 ± 0.26 ^a

Number of replicates, n = 2; Number of observations, N = 12; Mean value ± s.d.
^{a-d} values per column without common superscripts were significantly different ($p < 0.05$).

nozzle setup used in this study has limited processing capacity. In addition, future studies should quantify energy consumption, evaluate microbiological stability and powder-forming ability, and assess the functional properties of the concentrated or dried product after reconstitution.

5. Conclusions

The distance between the tip and the collector did not significantly affect droplet size, likely due to opposing effects between electric field intensity and residence time. It was also observed that a large tip diameter resulted in big drops, while a high fat content generated small drops due to the increase in viscosity and decrease in surface tension.

At sufficiently low flow rates (0.01 mL min^{-1}) with a voltage intensity of 29 kV and a tip-collector distance of 10 cm, electrospraying induced a very significant removal of water. Milk reached the greatest concentration factor ($Q > 3$) when initial milk solids concentration was the smallest, giving an inversely proportional relationship. This effect could be explained by the effect of increased solids concentration on physicochemical properties of the solution, preventing the evaporation of the solvent. In these processing conditions, electrospraying did not affect the structure of major milk proteins: caseins and whey proteins.

This study demonstrated, as a proof of concept, that electrospraying can promote partial water removal from reconstituted skimmed milk under specific low-flow-rate conditions, while no major changes in the SDS-PAGE profile of the main milk proteins were detected. Although a dried powder was not obtained, the results provide initial evidence that electrospraying may be used to concentrate milk without applying conventional thermal drying conditions.

CRediT authorship contribution statement

Bernat Perez-Playà: Writing – review & editing, Writing – original draft, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Anna Zamora:** Writing – review & editing, Validation, Supervision, Resources, Project administration, Methodology, Investigation, Funding acquisition. **Llibertat Abad:** Writing – review & editing, Supervision, Resources, Project administration, Methodology, Investigation, Funding acquisition. **Manuel Castillo:** Writing – review & editing, Validation, Supervision, Software, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledges

Supported by AGAUR (FI SDUR 2021 00324, SGR-2021-00497, SGR-2021-00618 and 2025 PROD 00093) and UAB Proof of Concept (PoC 2020); and Spanish ICTS Network MICRO-NANOFABS, partially funded by FEDER funds through MINATEC-PLUS-2 project FICTS2019-02-40, and CEX2023-001397-M financed by MICIU/AEI/10.13039/501100011033.

Data availability

Data will be made available on request.

References

- Barrero, A., & Loscertales, I. G. (2007). Micro- and nanoparticles via capillary flows. *Annual Review of Fluid Mechanics*, 39(1), 89–106. <https://doi.org/10.1146/annurev.fluid.39.050905.110245>
- Bhushani, J. A., & Anandharamakrishnan, C. (2014). Electrospraying and electrospraying techniques: Potential food based applications. *Trends in Food Science and Technology*, 38(1), 21–33. <https://doi.org/10.1016/j.tifs.2014.03.004>
- Bocci, D., & Casas, M. (2013). *Producción de Leche en Polvo Entera, Parcialmente Descremada y Descremada*. [Final Project, Universidad Nacional de Cuyo]. Biblioteca Digital Uncuyo. <https://bdigital.uncu.edu.ar/7878>
- Bohr, A., Boetker, J. P., Rades, T., & Yang, M. (2013). Application of spray-drying and electrospraying/electrospinning for poorly watersoluble drugs: A particle engineering approach. *Current Pharmaceutical Design*, 20(3), 325–348. <https://doi.org/10.2174/13816128113199990399>
- Čarić, M., Akkerman, J. C., Milanović, S., Kentish, S. E., & Tamime, A. Y. (2009). Technology of evaporators, membrane processing and dryers. In A. Y. Tamime (Ed.), *Dairy powders and concentrated products* (pp. 99–148). Wiley-Blackwell. <https://doi.org/10.1002/9781444322729.ch3>
- Deshwal, G. K., Akshith, F. N. U., Kadyan, S., Sharma, H., Singh, A. K., Panjagari, N. R., & Meena, G. S. (2021). Applications of reverse osmosis in dairy processing: An Indian perspective. *Journal of Food Science and Technology*, 58, 3676–3688. <https://doi.org/10.1007/s13197-020-04958-6>
- Dhiman, A., Suhag, R., Singh, A., & Prabhakar, P. K. (2022). Mechanistic understanding and potential application of electrospraying in food processing: A review. *Critical Reviews in Food Science and Nutrition*, 62(30), 8288–8306. <https://doi.org/10.1080/10408398.2021.1926907>
- Eskin, M., & Goff, D. (2013). Milk. In M. Eskin, & F. Shahidi (Eds.), *Biochemistry of foods* (3rd ed., pp. 187–214). Academic Press.
- Fox, P., Uniacke-Lowe, T., McSweeney, P., & O'Mahony, J. (2015). Heat-induced changes in milk. In P. Fox, T. Uniacke-Lowe, P. McSweeney, & J. O'Mahony (Eds.), *Dairy chemistry and biochemistry* (2nd ed., pp. 345–376). Springer International Publishing.
- Freire, P. A. (2022). *Synchronous front-face fluorescence of milk and whey as an analytical tool for assessing technological functionality of whey*. doctoral thesis. Universitat Autònoma de Barcelona. Tesis Doctorals en Xarxa <http://hdl.handle.net/10803/687705>.
- Ghorbani, M., & Santos, R. (2021). Electro-spinning and electro-spraying as innovative approaches in developing of a suitable food vehicle for polyphenols-based functional ingredients. In L. Queiroz, T. Casagrande, & E. Jacob-Lopes (Eds.), *Bioactive compounds – Biosynthesis, Characterization and Applications*. IntechOpen. <https://doi.org/10.5772/intechopen.77427>
- Gomez, A., & Deng, W. (2011). Fundamentals of cone-jet electrospray. In *Aerosol Measurement: Principles, Techniques, and Applications* (3rd ed., pp. 435–448). Wiley.
- Hartman, R. P. A., Brunner, D. J., Camelot, D. M. A., Marijnissen, J. C. M., & Scarlett, B. (1999). Electrohydrodynamic atomization in the cone-jet mode physical modeling of the liquid cone and jet. *Journal of Aerosol Science*, 30(7), 823–849. [https://doi.org/10.1016/S0021-8502\(99\)00033-6](https://doi.org/10.1016/S0021-8502(99)00033-6)
- Hartman, R. P. A., Brunner, D. J., Camelot, D. M. A., Marijnissen, J. C. M., & Scarlett, B. (2000). Jet break-up in electrohydrodynamic atomization in the cone-jet mode. *Journal of Aerosol Science*, 31(1), 65–95. [https://doi.org/10.1016/S0021-8502\(99\)00034-8](https://doi.org/10.1016/S0021-8502(99)00034-8)
- Hernández, C. (2018). *Análisis de distribuciones de carga espacial emitidas por atomización electrohidrodinámica (electrospray) en vacío*. Doctoral thesis. Universidad Nacional de Educación a Distancia. DSpace <https://e-spacio.uned.es/entities/publication/e6207f78-bdf5-44d4-a791-c769c2a853e1>
- Jafari-Nodoushan, M., Barzin, J., & Mobedi, H. (2015). Size and morphology controlling of PLGA microparticles produced by electro hydrodynamic atomization. *Polymers for Advanced Technologies*, 26(5), 502–513. <https://doi.org/10.1002/pat.3480>
- Jiang, Z., & Yu, X. (2018). Understanding of droplet dynamics and deposition area in electrospraying process. *Applied Physics*. <https://doi.org/10.48550/arXiv.1809.09688>. Article 1809.09688.
- Jovanovic, S., Barac, M., Macej, O., Vucic, T., & Lacnjevac, C. (2007). SDS-page analysis of soluble proteins in reconstituted milk exposed to different heat treatments. *Sensors*, 7(3), 371–383. <https://doi.org/10.3390/s7030371>
- Kumar, P., Sharma, N., Ranjan, R., Kumar, S., Bhat, Z. F., & Jeong, D. K. (2013). Perspective of membrane technology in dairy industry: A review. *Asian-Australasian Journal of Animal Sciences*, 26(9), 1347–1358. <https://doi.org/10.5713/ajas.2013.13082>
- Laemmli, U. (1970). Cleavage of structural proteins during the assembly of the head of bacteriophage T4. *Nature*, 227, 680–685. <https://doi.org/10.1038/227680a0>
- Landa, G. (2017). *Desarrollo de andamios fibrosos de policaprolactona, condroitín sulfato y ácido hialurónico para regeneración de cartilago*. Master's thesis. Universidad de Zaragoza. Zeguan <https://zaguan.unizar.es/record/76326?ln=es>.
- Lara-Aguilar, S., & Alcaine, D. (2019). Lactose oxidase: A novel activator of the lactoperoxidase system in milk for improved shelf life. *Journal of Dairy Science*, 102(3), 1933–1942. <https://doi.org/10.3168/jds.2018-15537>
- Man, Y., Zhou, C., Adhikari, B., Wang, Y., Xu, T., & Wang, B. (2022). High voltage electrohydrodynamic atomization of bovine lactoferrin and its encapsulation behaviors in sodium alginate. *Journal of Food Engineering*, 317, Article 110842. <https://doi.org/10.1016/j.jfoodeng.2021.110842>
- Mari, A., Nikoleta Parisouli, D., & Krokida, M. (2024). Exploring osmotic dehydration for food preservation: Methods, modelling, and modern applications. *Foods*, 13(17), 2783. <https://doi.org/10.3390/foods13172783>
- Moreira, A., Lawson, D., Onyekuru, L., Dziemidowicz, K., Angkawitwong, U., Costa, P. F., ... Williams, G. R. (2021). Protein encapsulation by electrospinning and

- electrospraying. *Journal of Controlled Release*, 329, 1172–1197. <https://doi.org/10.1016/j.jconrel.2020.10.046>
- Mukherjee, N., Bansal, B., & Chen, X. D. (2005). Measurement of surface tension of homogenised milks. *International Journal of Food Engineering*, 1(2), 2. <https://doi.org/10.2202/1556-3758.1013>
- Patil, M. H., Tanguy, G., Le Floch-Fouéré, C., Jeantet, R., & Murphy, E. G. (2021). Energy usage in the manufacture of dairy powders: Advances in conventional processing and disruptive technologies. *Drying Technology*, 39(11), 1595–1613. <https://doi.org/10.1080/07373937.2021.1903489>
- Persano, L., Camposo, A., Tekmen, C., & Pisignano, D. (2013). Industrial upscaling of electrospinning and applications of polymer nanofibers: A review. *Macromolecular Materials and Engineering*, 298(5), 504–520. <https://doi.org/10.1002/mame.201200290>
- Ratti, C. (2001). Hot air and freeze-drying of high-value foods: A review. *Journal of Food Engineering*, 49(4), 311–319. [https://doi.org/10.1016/S0260-8774\(00\)00228-4](https://doi.org/10.1016/S0260-8774(00)00228-4)
- Rayleigh, L. (1882). On the equilibrium of liquid conducting masses charged with electricity. *Philosophical Magazine Series*, 14(87), 184–186. <https://doi.org/10.1080/14786448208628425>
- Rebollo, N. (2015). *Análisis experimental de procesos de atomización de líquidos basados en campos hidrodinámicos y electrohidrodinámicos*. Doctoral thesis. Universidad de Extremadura. DEHESA <http://hdl.handle.net/10662/3919>.
- Schuck, P. (2009). Effects of drying on milk proteins. In A. Thompson, M. Boland, & H. Singh (Eds.), *Milk proteins: From expression to food* (3rd ed., pp. 283–306). Academic Press.
- Smith, A. (2015). *Electrohydrodynamic atomization produced nanoparticles for the targeted delivery of cancer chemotherapeutics*. Doctoral thesis. University of Portsmouth. Pure.
- Tro, N. (2022). Liquids, solids, and intermolecular forces. In N. Tro, T. Fridgen, & L. Shaw (Eds.), *Chemistry: A molecular approach* (3rd ed., pp. 418–478). Pearson.
- Walstra, P., Wouters, J., & Geurts, T. (2006a). Concentration process. In *Dairy science and technology* (2nd ed., pp. 297–336). CRC Press.
- Walstra, P., Wouters, J., & Geurts, T. (2006b). Appendix A.11. In *Dairy science and technology* (2nd ed., pp. 758–761). CRC Press.
- Whetstone, M. E. C., & Drake, M. (2007). The flavor and flavor stability of skim and whole milk powders. In K. R. Cadwallader, M. Drake, & J. McGorin (Eds.), *Vol. 971. Flavor of dairy products* (pp. 217–251). ACS Symposium Series.
- Williams, A. M., Jones, J. R., Paterson, A. H. J., & Pearce, D. L. (2005). Milks and milk concentrates: Surface tension measurement. *International Journal of Food Engineering*, 1(1), 1. <https://doi.org/10.2202/1556-3758.1002>
- Xie, J., Jiang, J., Davoodi, P., Srinivasan, M., & Wang, C. (2014). Electrohydrodynamic atomization: A two-decade effort to produce and process micro-/nanoparticulate materials. *Chemical Engineering Science*, 125, 32–57. <https://doi.org/10.1016/j.ces.2014.08.061>
- Xie, J., & Wang, C.-H. (2007). Encapsulation of proteins in biodegradable polymeric microparticles using electrospray in the Taylor cone-jet mode. *Biotechnology and Bioengineering*, 97(5), 1278–1290. <https://doi.org/10.1002/bit.21334>
- Zhang, Y., Liu, C., Lin, Y., Wu, H., Lin, Y., Zhao, F., & Han, S. (2024). Development of food-grade xylanase encapsulation using electrospray dextran microparticles. *Food Hydrocolloids*, 150, Article 109716. <https://doi.org/10.1016/j.foodhyd.2023.109716>
- Zhou, H., & Biswas, P. (2021). The prediction of size and charge of particles formed from evaporation of charged droplets generated in an electrospray system. *Chemical Engineering Science*. <https://doi.org/10.1016/j.ces.2020.116237>, 116237.