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A review on electrospraying technique for encapsulation of nutraceuticals

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Abstract
Electrospraying is a process of liquid atomisation by electrical forces. It was highly charged and produces droplets, which prevents their coagulation and supports self-dispersion. This technique, also known as electro-hydrodynamic atomization has established its technique in food nanotechnology as an easy and efficient method. Electrosprayed particles having a structural and functional advantages and makes micro and nano particles by controlling some parameter such as adjusting the flow rate and voltage applied to the nozzle. So, Electrospraying technique can considered as a route of nanotechnology and it can be used for different purposes such as film coating, chocolate processing and stabilization of food ingredients, drug delivery, and especially encapsulation of nutraceuticals. This review provides basic idea and introduction about electrospraying process, principal mechanism and application in food and other industries.

Keywords: Electrospraying, electro-hydrodynamic, nanotechnology, drug delivery, nutraceuticals

Introduction
The area of food science and technology is of great interest in field of nanotechnology to develop the novel application in this area. Currently, nanotechnology has entered the food industry presenting its unique superiority and applications. There are several techniques utilized to prepare nanoencapsulated in the food industry and the advantages of this nanoparticles are to produce sub micron and nano size particles, high surface to volume ratio, minimize denaturation and sustained and controlled release, effective encapsulation, enhanced stability of bioactive and no thermal processed require [1]. The electrospraying technique has also known as electro-hydrodynamic atomization has establish its approach in food nanotechnology as an easy and effective method [12][4].
Electrospraying has been proposed as a process for nanoencapsulation in the addition of a high level of electrostatic force to minimize the size of the development of aerosol droplets to the nanoscale. The electrospray droplets size can range from hundreds micrometers down to some tens of nanometer. The droplets size distribution can be nearly monodisperse. Droplet generation and size can be controlled to some extent through the flow rate of the liquid and the voltage at the capillary nozzle. Electrospraying is used for micro and nano particle production, thin-film deposition and micro or nano-capsule formation. Electrospraying technique develops new drug delivery systems, medicine production, and nano-encapsulation of nutraceuticals.
The setup for electrospraying having mainly four components: (1) a high voltage source (1-30 kV), (2) stainless steel needle or capillary spinneret, (3) syringe pump and (4) a grounded collector either flat plate or rotating drum [13].

Principle electrospraying technique
Electrospraying is method of liquid atomization by electrical force. The theory of this method is based on the potential of an electric field to crush a droplet and transform them to the micro or nanometer scale and its depending on control parameter [29].
When electric field is applied to a drop an electric charge is created within the droplet named as Coulomb force. This force competes with the cohesive force of the particle when it dominates over the cohesive force a dimution in the surface tension occurs and ultimately nanoparticles are obtained [3]. One of the techniques to calculate the rupture of the drop is to calculate a parameter known as Rayleigh by the electrostatic force [27].

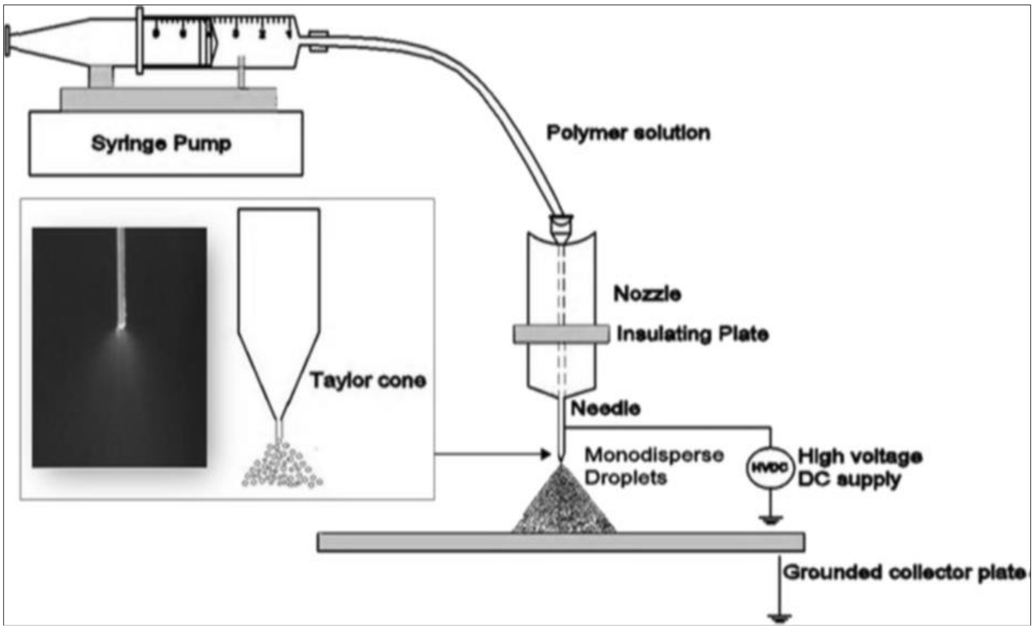

Fig 1: Schematic figure of electrospraying (Source: AnuBhushani *et al*., 2008) [1]

Stages of electrospraying atomization process (Tapia-Hernandez, Rodrıguez-Felix *et al*., 2017)

- First, the fluid must be dynamic with an initial acceleration so that the Taylor cone is formed. The Taylor cone is formed via a balance of forces as surface tension, gravity, liquid surface and electric tension, inertia, and viscous stress.
- Second, the fluid is ruptured within the cone jet forming tiny droplets.
- Last, the droplets are sprayed onto a collector surface.

As per data shown in table 1, electrospraying parameters such as equipment, solution and environmental condition are affect the particle size. A high electric potential and flow rate results a smaller particle diameter and large size particles and vice-versa. The large collector distance gives spherical morphology and more volatilization of solvent and short distance results in collapse of particle. A high concentration, viscosity and density form larger particles and vice-versa. Electrical conductivity is high to obtained smaller diameter and vice-versa. Relative humidity if ≤30% results smaller particles and vice-versa.

Table 1: Electrospraying parameters and their influence on the particle size

Parameters		Characteristics	
		Increases	**Decreases**
Equipment	Electric potential	high electrical potential (greater than 10 kv) Results in the formation of particles of smaller diameter	low electrical potential (less than 10 kv) There is no formation of Taylor cone or particles of larger diameter
	Flow rate	At a high flow rate,(≥ 1 mL/h)it can lead to the formation of large size particles(≥ 1µm)	At a low flow rate,(< 1 mL/h)and increasing the electrical conductivity can lead to smaller size particles(< 1µm)
	Collector distance	A large distance(15-20 cm or greater) provides more spherical morphology and more volatilization of solvent	Short distance(5-14 cm) results in collapse of the particle
Solution	Concentration	high concentration tend to form larger particles	low concentration tend to form smaller particles
	Viscosity	Increases if the concentration of the polymer is increased and larger particles are obtained	By increasing the voltage and the flow rate, the viscosity decreases and smaller particles are obtained
	Density	Thicker Taylor cone, larger inertial force, therefore droplets are larger	Smaller diameter Taylor cone decreasing fluid resistance, small particles are formed
	Electrical Conductivity	If is high, exceed surface tension and smaller particle diameter will be obtained	If is low, higher surface tension, generates larger particles
Environmental	Relative humidity	If ≤30%, all the solvent if volatilized and completely dry polymer reaches the collector, smaller particles are obtained	if >30%, no uniform particle diameter and not defined morphology, larger particles are obtained

Application of electrospraying technique

- Electrospraying technique can be manipulate for several purposes: mainly, it can be used in the area of inedible food, being used to develop intelligent packaging that can oppose the entrance of pathogens [10]. It was used to produce nanostructures based in food polymeric materials, that is, to create particles for encapsulation of bioactive substances or food ingredients [18, 19].
- Electrospraying technique can be used for various purposes, for example film coating, chocolate processing, and stabilization of food ingredients, drug delivery, and particularly encapsulation of bioactive and nutraceutical compounds [5]. It is applicated in the use of polymeric matrices for encapsulation of nutraceuticals [21]. This technique firstly used for encapsulating drugs for making medicine with the intend of extended and/or controlled release. A research performed by Lee *et al*. (2010) [14].

- By using chitosan coating on polyethylene (PE) as packaging material in food by electrospraying, give some level of protection to fruits and vegetables as decreased gas permeability and antibacterial properties [17].
- Electrospraying is used to manufacture fabricating edible films and coating to get better the safety and quality of foods. Pareta and Edirisinghe (2006) [20] elaborated edible films of corn starch by electrospraying. Electrospraying used to incorporated essential oils and pullulan nanoparticles in films in order to control hazardous bacteria in meat and poultry products [16].
- Coaxial electrospraying method can used to prepare and describe nanoparticles of folate–chitosan–gemcitabine, obtaining nanoparticles in the range of 200 to 300 nm with activity against pancreatic cancer [23].
- By using electrospraying to encapsulate curcumin in a gelatin network has been shown to be an effectual way to raise its water solubility, civilizing its dispersion/solubility in the aqueous food matrix used as a food model (a gellified fish product), in addition to its bioaccessibility [7]. Electrospraying technique employed to obtain food-grade gelatin capsules so as to guard bioactives [8].
- As per data shown in table 2, so many studies of nutraceutical encapsulated by electrospraying techniques in which nanoancapsulate folic acid using eletrospraying technique into the whey protein concentrate (WPC) matrix and commerical resistance starch resulting to produce smaller particle sizes compared to nanopsray drying Nanoencapsulation enhanced the bioavaibility and stability of folic acid That means this phenomena lies within the interaction between the folice acid and protein matrix which bolsters the stability [26].
- Zein is a corn protein and it was used as the polymer for the encapsulation of docosahexaenoic acid (DHA) and curcumin in order to get submicron (490±200 nm) and nanoparticles (175-250 nm) respectively. The chemical stability of encapsulated DHA increased against degradation, and improved stability to environmental conditions such as relative humidity and temperature and reduced off-flavour [22].
- Electrospraying techniques used to prepare whey protein concentrate (WPC) micro, sub micro and nanocapsules for the encapsulation of bioactives, applications in the interest in the development of novel functional foods. The capsules have also established to capable and become stable functional additives i.e., antioxidant β-carotene which was encapsulate or electrosprayed in WPC capsules containing glycerol. The technique has been reported incredibly high encapsulation efficiency and successfully developed the capsules and stabilized the antioxidant against photo-oxidation [15].

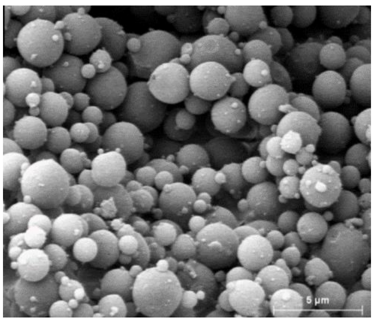

Fig 2: Electrosprayed 40% wt WPC aqueous solution (source: Lopez-Rubio & Lagaron, 2012) [15].

- Electrospraying technique used for microencapsulation of peppermint oil in alginate (A) and pectin (P) matrix, the microcapsules were prepared by mixing the necessary quantities of both polymers in distilled water to generate six different ratios of alginate to pectin (P), that is, 100:0, 80:20, 60:40, 40:60, 20:80 and 0:100 (%w/v) and characterized by determining their compositions and properties. The minimum size (1.58 µm) was obtained with A80: P20, while the maximum (3.24 µm) was obtained with A0: P100. The zeta potential value with all combination was ranged from 53.1 to 21.7 mV. The polydispersity index (PDI) tended to increase with the pectin content [24].
- To prepared ALA-loaded capsules via gelatin, whey protein concentrate, or soy protein isolate as carrier materials with help of electrospraying technique achieving microencapsulation efficiencies (MEE) is up to ~70% as compared to spray drying [8]. The capability of electrospraying of protein stabilized emulsions for the microencapsulation and superior protection of thermo-sensitive and hydrophobic bioactive ingredients, specially ω-3 fatty acids, offering an improved alternative to traditional technologies used in the food industry such as spray-drying, which gives increase the oxidative degradation and does not significantly protect ω-3 fatty acids [30].
- Gomez-Estaca *et al*. (2012) [6] investigated the effect of zein concentration on the morphology of the ensuing zein structures for polymer concentrations ranging from 1% to 20% (w/w) in aqueous ethanol solutions. It was found that 1% of zein in the solution was also low for particle formation, while a zein concentration of 20% gave rise to the transition from particles to fibers, a finding which was in accordance with observations made in earlier work. With zein concentrations of 2.5 and 5% (w/w) the generated particles were round in shape and had relatively smooth surfaces, whereas much larger particles in size were obtained by increasing the polymer concentration. A raise in the size of particle among higher polymer concentration and viscosity has been also noted for other polymers, such as polycaprolactone (PCL), alginate, chitosan and elastin.
- Xie *et al*. made-up a micron sized particles from a broad variety of polymers and using different electrospraying parameters with controllable morphologies [27]. A single cone jet was achieved when applying a voltage in between to nozzle and a ring placed in the path to the collector, For polycaprolactone (PCL) concentration between 6 to 0.5 wt% and voltage of 7-9 kV, the morphology of particle was changed from smooth to rough to corrugated and unequal shape with decreasing concentration along with decreasing particle size, even upto 300-600 nm at low flow rates. At similar voltage, the PCL particle size increased to 17-30 µm for increasing flow rates of 3-15 mL/h. Further, electrospraying (7-9 kV, 3 mL/h) of 3 wt% of PCL in solvents with varying conductivities/ dielectric constants (dichloromethane, tetrahydrofuran and acetonitrile) resulted in particle sizes inversely proportional to solution conductivity. Lastly, different shapes ranging from spheres to donut to corrugated shapes were possible by electrospraying of different types of polymers as PCL, poly (D, L-lactic-co-glycolic acid) (PLGA) and poly (L-lactide) (PLLA).

- A low-priced, low-energy and organic solvent-free encapsulation technology was studied by utilizing the pH-dependent solubility properties of curcumin and self assembly properties of sodium caseinate (NaCas). Curcumin was deprotonated and dissolved, while NaCas was dissociated at pH 12 and 21 C for 30 min. The subsequent neutralization enabled the encapsulation of curcumin in self-assembled casein nanoparticles. The degradation of curcumin under encapsulation conditions was negligible based on visible light and nuclear magnetic resonance spectroscopy. The dissociation of NaCas at pH 12 and reassociation after neutralization were confirmed using dynamic light scattering and analytical ultracentrifugation. The curcumin encapsulated in casein nanoparticles showed significantly improved anti-proliferation activity against human colorectal and pancreatic cancer cells. The studied encapsulation method is promising to utilize lipophilic compounds in food or pharmaceutical industries [24].
- In vitro analysis has shows sorghum condensed tannins (SCT) encapsulated in kafirin micro-particles can decrease blood glucose levels similar to acarbose, after ingestion of carbohydrate in healthy rats after an Oral starch tolerance test (OSTT). Sorghum condensed tannins -SCT-KEMS can also prevent elevation of serum insulin. By encapsulation of SCT in kafirin micro-particles, it seems to cover the bitterness and astringency of SCT and enables them to be delivered to the small intestine where they inhibit carbohydrate hydrolysis. These several effects exerted by SCT-KEMS are due to SCT's strong affinity for the proline-rich kafirin [31] and kafirin's slow digestibility by intestinal proteinases as consequence of its hydrophobicity and disulphide-bonded cross-linking [32]. Thus, encapsulating SCT in kafirin microparticles has likely as a novel, affordable nutraceutical-type action for the management of hyperglycaemia [28].
- The electrospraying was to optimize the fabrication parameters which included in for reproducible synthesis of chitosan based micro or nanospheres and to study their potential as delivery vehicles for bioactive agents. The analysis of SEM verified that microspheres of less than 1 µm were obtained when chitosan concentration was 2% dissolved in 90% acetic acid. The favourable results were working distance and needle gauge that yielded 7 cm and 26 g, respectively. Ampicillin loaded chitosan micro/nanospheres having average particle size of was 520 nm with zeta potential of 128.2 mV and 80.4% encapsulation efficiency. The particles were categorized for drug release kinetics and results verified an initial burst release followed by a sustained release over a period of 120 h. Further, antibacterial activity of drug loaded micro or nanospheres confirmed that the encapsulated drug was in its active form post exposure to high voltage during electrospraying. This study indicates that electrospraying is a facile technique for the synthesis of chitosan micro or nanospheres for drug delivery applications [2].

Table 2: Studies of nutraceutical encapsulation by electrospraying technique

Technique used	Wall Material Used	Core Material	Size (nm)	Purpose	Reference
Electrospraying	Zein ultrathin fibers	DHA	500-700	Improved stability	[22]
	Chitosan micro-/nanospheres	Ampicillin sodium	455-885	Higher encapsulation efficiency and improved anti-bacterial activity	[2]
	Whey protein concentrate	Carotene	<100	Whey protein explored as the coat material to deliver bioactives	[15]
	Zein	Curcumin	175-250	Enhanced stability and dispersion in aqueous food matrix	[6]
	Sodium Caseinate	Curcumin		Antiproliferation activity	[25]
	Gelatin, WPC and soybean protein	α-Linolenic acid		Nutraceuticals	[8]
	Alginate-pectin	Peppermint oil		Perfumes, cosmetics, medicines, and the flavoring industry	[24]
	Kafirins	Extract of sorghum condensed tannins		Inhibiting α amylase	[28]

Conclusion

Electrospraying is a flexible apparatus for atomization of liquid that has the benefit of generation uniform droplets from inexpensive equipment. Electrospraying is a single-step, low-energy and low-cost material processing technology for the production of encapsulation and delivery systems. Electrosprayed polymer particles can be used as delivery system nutrients to protect them during processing and storage or during passage of the components to the target site in body. There is clear potential to develop electrosprayed particles to improve the design and function of novel products and delivery systems for functional food compounds.

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