Mechanistic understanding and potential application of electrospraying in food processing: a review

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CRITICAL REVIEWS IN FOOD SCIENCE AND NUTRITION
https://doi.org/10.1080/10408398.2021.1926907

REVIEW

Mechanistic understanding and potential application of electrospraying in food processing: a review

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ABSTRACT
Electrospraying (ESPR) is a cost effective, flexible, and facile method that has been used in the pharmaceutical industry, and thanks to its wide variety of uses such as bioactive compound encapsulation, micronization, and food product coating, which have received a great attention in the food market. It uses a jet of polymer solution for processing food and food-derived products. Droplet size can be extremely small up to nanometers and can be regulated by altering applied voltage and flow rate. Compared to conventional techniques, it is simple, cost effective, uses less solvent and products are obtained in one step with a very high encapsulation efficiency (EE). Encapsulation provided using it protects bioactives from moisture, thermal, oxidative, and mechanical stresses, and thus provides them a good storage stability which will help in increasing the application of these ingredients in food formulation. This technique has an enormous potential for increasing the shelf life of fruit and vegetables through coating and improvement of eating quality. This study is aimed at overviewing the operating principles of ESPR, working parameters, applications, and advantages in the food sector. The article also covers new ESPR techniques like supercritical assisted ESPR and ESPR assisted by pressurized gas (EAPG) which have high yield as compared to conventional ESPR. This article is enriched with good information for research and development in ESPR techniques for development of novel foods.

KEYWORDS
Electrospraying; coating; encapsulation; film formation; food applications

Introduction

Electrospraying (ESPR) technique involves formation of small and uniform-sized particles or thin film coating by electrostatically accelerating the material to break it into micro or nanosized particles (Doyle et al. 2013). ESPR uses "bottom up" approach to create micro and nanoparticles (Lim 2015). In the field of food processing, ESPR can find application in micro and nano particle production, food coating, film formation, electro-emulsification, and electro-encapsulation (Drosou, Krokida, and Biliaderis 2017; Jaworek and Sobczyk 2008). Different types of ESPR systems are ESPR on plate or conventional ESPR, emulsion ESPR, core–shell ESPR, and coaxial ESPR (Kurakula and Raghavendra Naveen 2021). For size and morphology of the final product, factors such as polymers concentration, viscosity, and molecular weight and process parameters such as distance between needle tip and collector, flow rates, applied voltage and flow rates, etc., are significant. By manipulating these parameters, morphologies like nanocups, porous microcarriers, janus particles, core-wall/multi-layered microspheres, and cell-shaped particles are obtained (Wang, Jansen, Yang 2019). ESPR has been applied in microencapsulating bioactive compounds. Microencapsulation and nanoencapsulation is a process in which an active component is packed inside a protective material which acts as a functional barrier to avoid any chemical and physical change in the properties of the active component (Bakry et al. 2016). Bioactives like vitamins, antioxidants, phenolics, and probiotics are used for food fortification but these have low stability thus to avoid deterioration it requires appropriate processing technique (Jacobsen et al. 2018). Through encapsulation, these bioactives are protected against oxidation, thermal stress, moisture, and their shelf life are enhanced. Another purpose of encapsulating active components is to facilitate its delivery, like conversion of liquid into small dried particles simplifies the handling process. Techniques like spray drying, coacervation, freeze-drying, melt extrusion, and ESPR have been used to microencapsulate products like vegetable oils (linseed oil, soybean oil, and olive oil), marine (fish oil and microalgal oil), and essential oils, etc. (Lim 2015; Walia et al. 2019). Soy protein isolate, casein, zein, whey protein concentrate (WPC), amylopectin, chitosan, guar gum, dextrin, polyethylene glycol, and polylactic acid are common protein, carbohydrate, and fat-based encapsulating materials used in encapsulation by ESPR (Alehosseini et al. 2018). Furthermore, ESPR can be used to entrap biomacromolecules such as enzymes, DNA, RNA, etc., without impairing their biological activity (Boda, Li, and Xie 2018). ESPR has been used for encapsulating food

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bioactives like anthocyanin extract, resveratrol, green tea catechins, curcumin, β carotene, etc., and probiotics like *Lactobacillus plantarum*, *Lactobacillus casei*, *Bifidobacterium lactis,* etc. (Wang et al. 2020).

Edible coatings act as a barrier to reduce quality deterioration process in foods by acting as a natural skin layer and preventing oxidation, moisture, and aroma loss along with making its mechanical handling easy. In addition to this, coatings may also act as a carrier of functional ingredients. ESPR has its application in forming ultra-thin and uniform edible liquid or powder coating which has been found to better than conventional methods of coating (Khan, Nazir, and Maan 2017). ESPR systems are also used for ionization purpose in combination with mass spectrometry to identify the unknown biomolecules (Kavadiya and Biswas 2018). Conventional ionization techniques like chemical and electronic ionization led to analyte fragmentation and structural destruction especially of thermolabile compounds. In the case of ionization by ESPR, multiple charged gas-phase ions are produced without any changes in native structures of thermally labile macromolecules like nucleic acid and proteins (Banerjee and Mazumdar 2012; Smith and Thakur 2010). Both high-performance liquid chromatography (Smith and Thakur 2010) and gas chromatography (Brenner et al. 2008) can be coupled to it. ESPR has also been used for micronization (particle size <10 μm) where reduction in particle size of food matrix leads to structural, rheological, functional, and physicochemical changes providing a processing advantage along with preserving it (Dhiman and Prabhakar 2021). Low throughput is one disadvantage of ESPR technique. ESPR assisted by pressurized gas (EAPG) is a novel approach of ESPR in which very high yield is obtained as compared to conventional ESPR, with similar advantages (Bioincia n.d.). Green chemistry involves use of certain set of principles which deals with reducing or eliminating formation of hazardous chemical substances in manufacturing processes. It targets delivering active compound with the highest therapeutic effect, efficiency, robustness, and safety by using certain set of techniques in order to reduce toxicity, improve performance and eliminate environmental pollution which reduces economic burden as well. Supercritical fluid (SCF) like supercritical CO_2 ($scCO_2$) are economical, nontoxic, non-inflammable, non-mutagenic, non-cancerous, and thermodynamically stable thus it is considered as a "green technology." Using SCFs along with ESPR can provide higher loading capacity along with reducing chances of analyte loss and activity of encapsulated compound (Demirdöğen et al. 2018; Dhiman and Prabhakar 2021). Using $scCO_2$ along with ESPR improves the process and production rate up to hundred times and opens channel for "green ESPR" (Baldino, Cardea, and Reverchon 2019b). ESPR's "green" consciousness and non-thermal approach, makes it a technology of huge interest and has immense potential opportunities for research and development on food matrix (Baldino, Cardea, and Reverchon 2019b; Soares et al. 2018). ESPR has applications in 2D, 3D, and direct printing as well (Jaworek, Sobczyk, and Krupa 2018). Electrohydrodynamic jet printing (Tanhaei et al. 2021) is a novel method of printing where instead of acoustic or thermal energy, electric fields are used to produce fluid flow needed to print patterns on a substrate. To the best of our understanding, it is still not used in food processing. The current review aims to highlight the potential of ESPR in food processing and further its working principle, parameters, advantages, and disadvantages have been discussed.

Mechanistic understanding of fundamentals of electrospraying

The study of the development of electrically charged fluids due to surface and volumetric forces induced by an electric permittivity gradient and/or a non-zero space charge density is known as electrohydrodynamics (Gañán-Calvo et al. 2018). ESPR is an electrohydrodynamically processing technique in which electric force is used to atomize the liquid and thus form extremely small droplets ranging from micro to nanoscale. The apparatus consists of high voltage supply (1–30 kV) which is connected to a capillary nozzle (Figure 1a) and grounded collector. The material to be atomized is passed through the nozzle where it gets elongated due to the electric field generated by voltage supply connected to the nozzle and is disrupted into small droplets due to the electric forces experienced by it (Bhushani and Anandharamakrishnan 2014; Jaworek and Sobczyk 2008). Solidification of particles during ESPR (Figure 2b) occurs due to two processes that are evaporation of solvent from the polymer solution boundary and simultaneous diffusion of solvent from core to the boundary of the polymer feed solution jet (Tanhaei et al. 2021). Microdroplets get charged in the electric field and are thus self-dispersing. Due to the charge present on them, they repel each other helping in preventing droplet coagulation (Bhushani and Anandharamakrishnan 2014; Jaworek and Sobczyk 2008). Charges developed on the droplets surpass the surface tension of the liquid and split the jet into fine and even droplets. The fluid that passes via the nozzle also experiences shear stress provided by the electrostatic forces which help in forming fine cone jets (Alehosseini et al. 2018). Flat-ended capillary, sawtooth capillary, tilted capillary, capillary with dielectric porous fiber, trumpet-shaped capillary, capillary with metal needle, and capillary with hairpin electrode are various types of nozzles (Figure 3a–h) used in ESPR equipment. In addition to these coaxial nozzle, double nozzle, dual nozzle, three coaxial nozzle, and triple nozzle (Figure 3i–m) are the nozzles used in ESPR (Jaworek, Sobczyk, and Krupa 2018). Various spraying modes are achievable in ESPR like micro dripping, spindle dripping, cone jet, and multi jets (Alehosseini et al. 2018). In the case of dripping mode (occurs at low voltage), fragments of liquid are expelled in elongated form of large droplets having diameter greater than nozzle diameter. Whereas in case of jet mode, spray of highly charged droplets ranging from micron to nano-level (disintegration due to high voltage) is emitted in form of steady liquid jet (Wang et al. 2019). Out of these, cone jet mode is most important and stable which provides smaller size droplets with narrow particle size

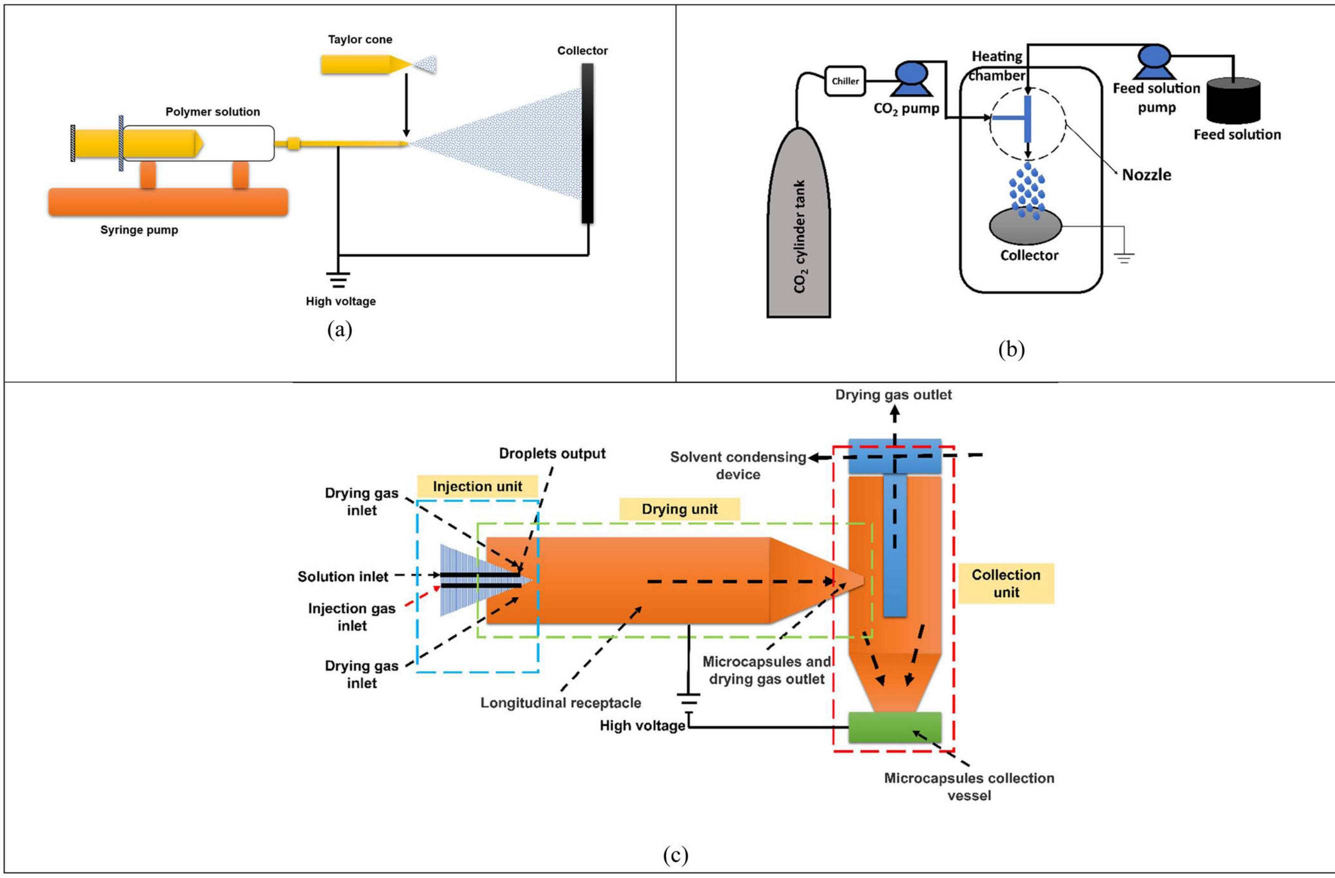

Figure 1. Schematic diagram of (a) electrospraying and (b) supercritical assisted electrospraying process (c) electrospraying assisted by pressurized gas (Adapted fromBaldino, Cardea, and Reverchon 2019b; Bhushani and Anandharamakrishnan 2014; Lagaron et al. 2018).

distribution (Alehosseini et al. 2018). Steady Taylor cone-jets are formed when the flow rate of liquid releasing from jet equals to the one feeding the conical meniscus (Gañán-Calvo et al. 2018). Taylor cones formation also involves balance of forces like gravity, liquid surface, surface tension, and inertia, as well as viscous stress and electric tension (Tapia-Hernández, Rodríguez-Félix, and Katouzian 2017). Processing fluid gets ruptured inside the cone jet leading to formation of small droplets (Raval et al. 2019). The bulk forces acting on the jet are electrohydrodynamic force, gravitational force, inertial force, and drag force, while normal and tangential stresses form at the liquid surface (Figure 2a). These bulk forces and stresses are responsible for droplet disruption and deformation (Jaworek and Sobczyk 2008). The collection of these droplets is commonly done on a flat collector or on a cross linking/oppositely charged solution like $CaCl_2$ which facilitates the formation of micro and nano-particles (Tapia-Hernández et al. 2015). An ideal cross-linking agent used should be nontoxic, biocompatible, low molecular weight, and should be miscible (Tapia-Hernández, Rodríguez-Félix, and Katouzian 2017). The morphology of the produced particles is very much dependent on the physicochemical properties of the solvent along with the ambient conditions, so solvent selection is also an important step toward the production of ESPR particles (Tanhaei et al. 2021). Two main types of microcapsules formed using ESPR are particle/matrix microcapsule (where matrix is formed by continuous phase and it uniformly entraps dispersed phase in for of microdroplets) and core/shell microcapsule (where the shell material encloses the continuous phase in form of droplets forming the core) (Jaworek, Sobczyk, and Krupa 2018). Morphologies like core-wall/multi-layered microspheres, janus particles, porous microcarriers, nanocups, hollow microspheres, and cell-shaped particles (Figure 4a) are obtained in encapsulation process. In case of coating porous (grainy, fractal-like, reticular) and dense (crystalline, amorphous, amorphous with intrusions) morphologies (Figure 4b) are obtained (Jaworek, Sobczyk, and Krupa 2018; Wang, Jansen, Yang 2019). The relationship between morphology of particle and the solidification processes (polymer diffusion and solvent evaporation) (Lee et al. 2018) is outlined by the equation no. 1

$$P = R_r \times \frac{r}{D} \dots \quad (1)$$

where R_r is the rate of change of droplet radius due to solvent evaporation, r is the droplet radius, and D is diffusion coefficient of solute inside the droplet.

In ESPR process, polymer used as a protective layer for food bioactives should be nontoxic, biodegradable, and food grade and should have good solubility in nontoxic solvents. In food industries, renewable biopolymers like chitosan, alginate, starch, and gums are used as wall material (Soares et al. 2018). Processing polysaccharides (e.g., alginate, starch, pullulan, dextran, modified celluloses, chitosan, and xanthan)

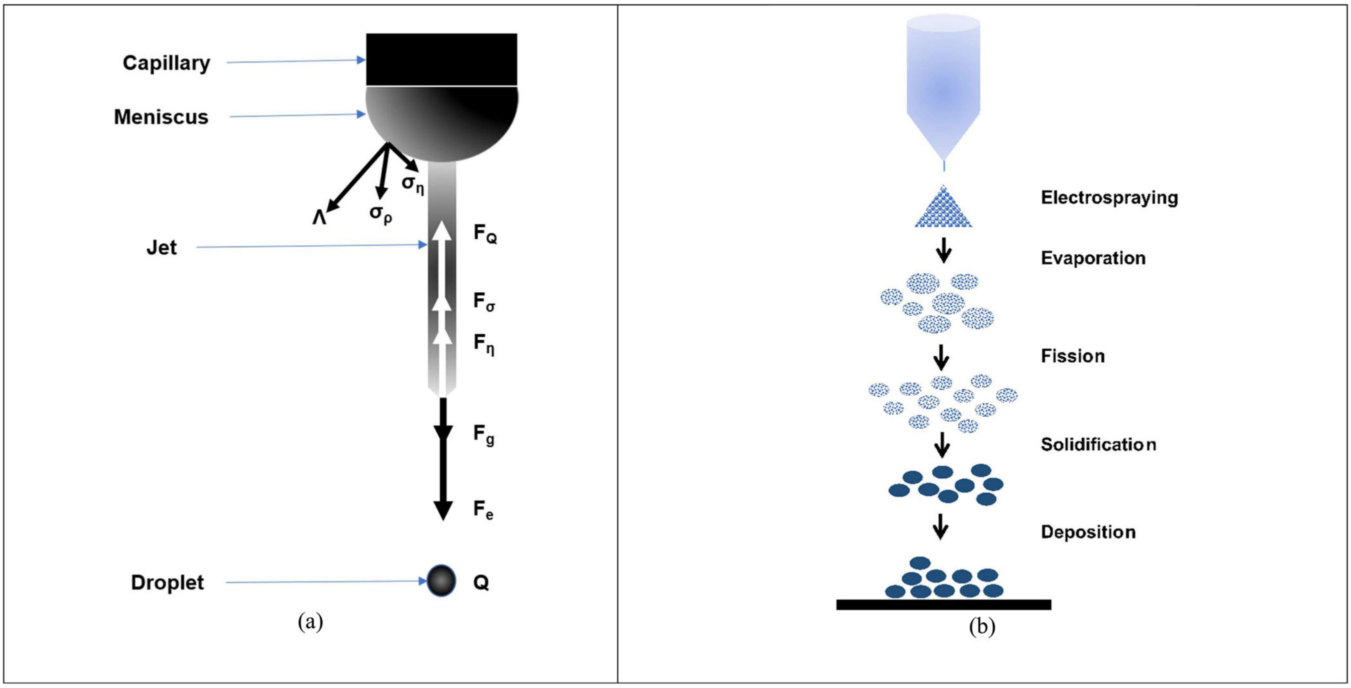

Figure 2. (a) Stresses and forces on electrohydrodynamic liquid jet, where Fe is electrohydrodynamic force, F_g is gravitational force, F_η is drag force, F_σ is inertial force, F_Q space charge of any previously emitted droplets, Q is surface charge density, Λ is local electric field, σ_η is the gradient of liquid velocity normal to inter-phase surface and σ_ρ is local velocity at the inter-phase surface (b) Production of micron and nano particles using electrospraying drying method (Adapted from Alehosseini et al. 2018; Jaworek, Sobczyk, and Krupa 2018).

using ESPR rely on their functional groups, degree of modification, and molecular weight. Whereas feasibility of processing proteins (zein, wheat gluten, whey proteins, soy proteins, and gelatin) using ESPR depends on its physicochemical, functional, and conformational properties (Lim 2021).

Similarly, EPSR ionization used in mass spectrometry consists of a stainless-steel capillary or hypodermic needle to which a dilute analyte solution is injected. At the tip of the capillary a high voltage of 2–6 kV is applied which leads to formation of taylor cone and disrupts the sample solution into aerosol of highly charged droplets due to coulombic explosion (Banerjee and Mazumdar 2012; Smith and Thakur 2010). ESPR coating also works in the same way; an electric field is applied on the coating material passing through the nozzle. Charges developed on liquid overcome its surface tension and apply shear stress on it breaking it into micro droplets. Droplets transfer their charge to the conductive target (that is the food material), initiating deposition. Coating material (no charge) starts filling the pores and then develops a thin layer on target material. In coating, ESPR has the advantage of depositing droplets on all the borders of target material called the "wrap around effect." Droplets appear to lose their charge, allowing them to cover as much of the target material as possible. This effect is influenced by the target surface conductivity and charge to mass ratio. The greater the charge-to-mass ratio and target surface conductivity, the greater the target material coverage (Khan, Nazir, and Maan 2017). Particle size and morphology of material is highly dependent on characteristics of solution, processing conditions, and environmental conditions, all of which are discussed in more detail in subsequent parts.

In ESPR, the droplet size depends on the process as well as fluid parameters (Table 1). In order to convert the liquid into tiny droplets, the applied voltage (V) (Equation (4)) should be such that it exceeds the Rayleigh limit i.e., the amount of electrostatic force (Equations (2 and 3)) that overcomes the fluid's surface tension split it into small droplets. If the surface tension is high, larger particles are formed. At very high-applied voltage, cone jets get unstable. Optimum range of electric potential is around 10–20 kV. Keeping flow rate of feed high leads to less solvent evaporation and thus forming large particles. At low flow rate, micro and nano particles up to 80 nm size are developed due to high volatilization of solvent (Khan, Nazir, and Maan 2017; Tapia-Hernández et al. 2015).

$$L = q\left(64\pi^2 \varepsilon \gamma R^3\right)\ldots \tag{2}$$

$$q = 8\pi\sqrt{\varepsilon_\circ \gamma R^3}\ldots \tag{3}$$

$$V = k\left[\gamma^2 K^2 \rho / {\varepsilon_\circ}^2\right]^{1/3}\ldots \tag{4}$$

L=Rayleigh charge limit, q=electrostatic charge, ε=liquid permittivity, $\varepsilon_\circ$=vacuum permittivity, γ=liquids surface tension, and R=droplet radius. V=required voltage, K=electrical conductivity, ρ =density, and k=constant in the order of unity.

The relationship between particle size of electrosprayed particle and flow rate, dielectric constant, droplet diameter, and liquid's conductivity (Niu et al. 2020b) is given by equation

$$d_p = \phi^{1/3} \times d \times G \times \varepsilon \times (Q \times \varepsilon \times \varepsilon_\circ / k)^{1/3}\ldots \tag{5}$$

where d_p=diameter of particle and d=diameter of droplet,

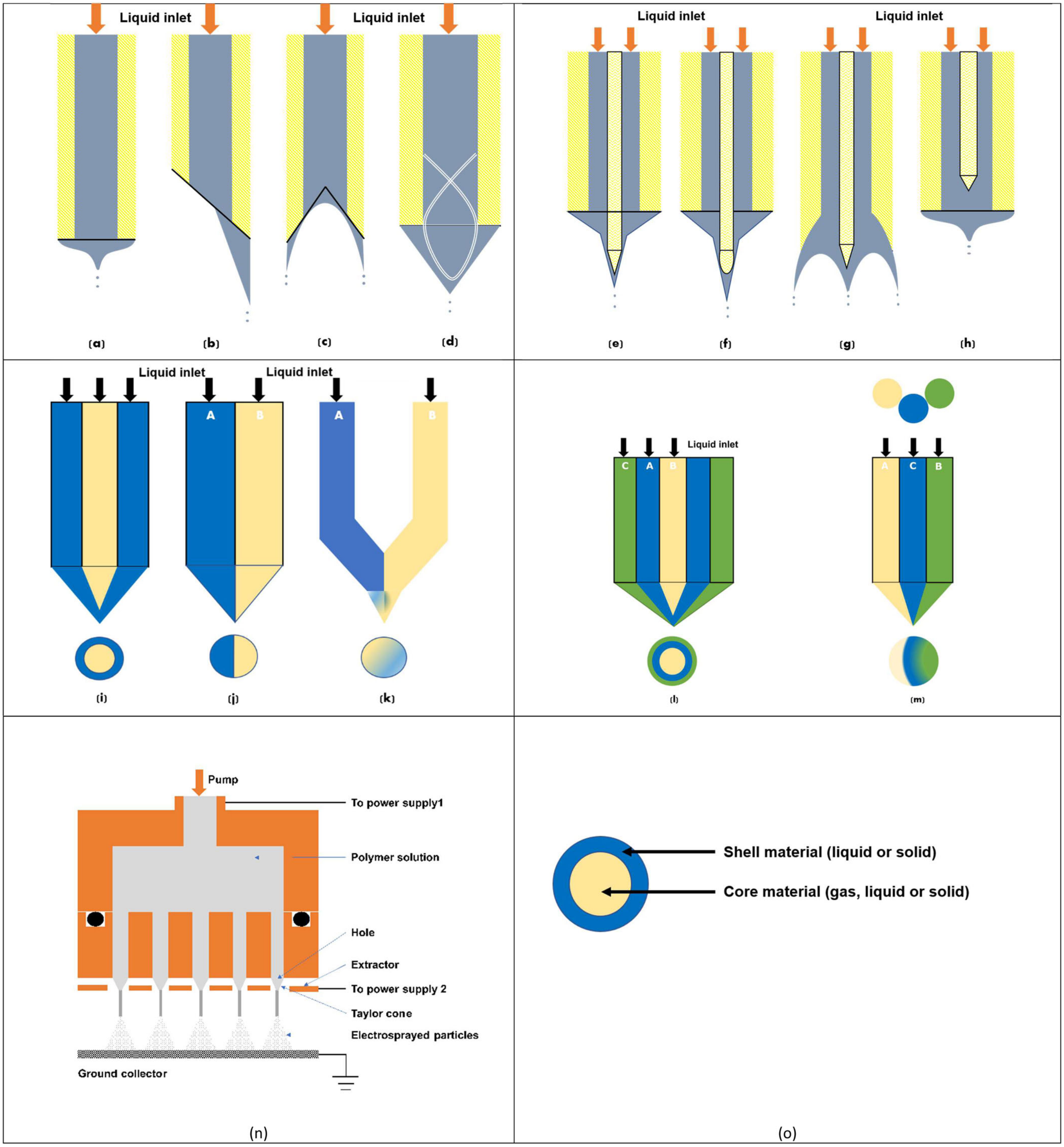

Figure 3. Schematic diagram of types of electrospray nozzles (a) flat-ended capillary, (b) tilted capillary, (c) sawtooth capillary, (d) capillary with hairpin electrode, (e) capillary with metal needle, (f) capillary with dielectric porous fiber, (g) trumpet-shaped capillary with an axial metal needle, (h) glass capillary with a metal ion injector, (i) co axial nozzle made of two coaxial capillaries (core shell matrix), (j) double nozzle made of two parallel capillaries (janus particles), (k) dual nozzle made of two capillaries with common outlet (particle/matrix microcapsule), (l) three coaxial nozzle made of three coaxial capillaries (core/double shell microcapsules), (m) triple nozzle of three parallel capillaries (three color janus particles), (n) atomizer set up for producing multiple jets and (o) core/shell microcapsule (Adapted from Jaworek, Sobczyk, and Krupa 2018; Lim 2015).

ϕ=polymer volume fraction, Q=flow rate of liquid, G=dielectric constant, k =conductivity of liquid, ε =liquid permittivity, and $\varepsilon_\circ$ = vacuum permittivity.

Tip to collector distance of 15–20 cm is considered as optimum for effective volatilization and formation of smaller droplets. Viscosity is another parameter which influences the particle size of droplets as flow rate and concentration are closely related to viscosity. The viscosity of the solution increases as the active component concentration increases, resulting in larger particles. Smaller particles are obtained at low viscosity. Electrical conductivity of material to be electrosprayed is another important parameter to form smaller particles without aggregation. Material with high conductivity gets charged quickly and experiences high Columbian

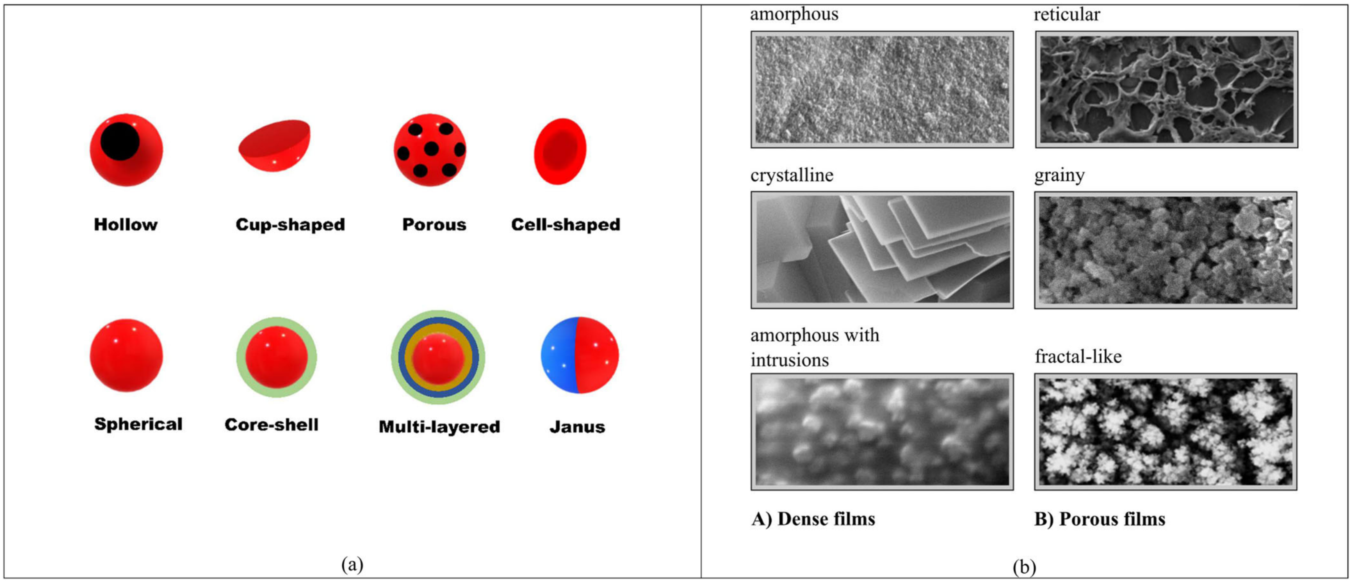

Figure 4. Schematic illustration of (a) morphology of various types of encapsulated particles produced by electrospraying (Adapted fromWang, Jansen, Yang 2019) and (b) morphology of various types of coatings produced by electrospraying (Adapted from Jaworek, Sobczyk, and Krupa 2018).

Table 1. Processing parameters in ESPR process.

S. no.	Properties		References
1.	Solution properties	Surface tension Electrical conductivity Polymer concentration Viscosity Molecular weight of polymer Contact angle	Alehosseini et al. (2018), Drosou, Krokida, and Biliaderis (2017), Khan, Nazir, and Maan (2017), Wang, Jansen, Yang (2019)
2.	Processing conditions	Applied voltage (1–20 kV) Tip to collector distance (7–15 cm) Flow rate (1–15 mL/h) Diameter of nozzle Solvent evaporation rate	
3.	Ambient conditions	Humidity Temperature	

repulsion forces and overcomes cohesive forces of material leading to smaller droplets formation (Drosou, Krokida, and Biliaderis 2017; Tapia-Hernández et al. 2015). Contact angle is also a critical parameter when it comes to coating. Contact angle means interactive surface behavior that is measure of hydrophilicity or lipophilicity of material. When the sprayed material and the material to be coated both are of same nature either hydrophilic or hydrophobic, it leads to fusion of sprayed particles leading to formation of larger particles (Khan, Nazir, and Maan 2017). Diameter of the nozzle is also an important factor, smaller the diameter smaller the particle size. But these nozzles get blocked on passing polymers with high glass transition temperature. Morphology and productivity of ESPR is also influenced by ambient parameters especially temperature and humidity because volatilization of solvent is affected by these conditions (Alehosseini et al. 2018). Therefore, it is better to perform ESPR process under controlled ambient conditions. Morphology of final product is also dependent on the solvent evaporation rate. Evaporation of solvent at slower rate leads to formation of solvent free polymer with stable spherical morphology whereas faster evaporation leads to particle fragmentation and forms poor quality droplets (Wang et al. 2020).

Different configurations of electrospraying process

Coaxial design of ESPR is a very novel and effective technology in microencapsulating core material with smaller droplet size. Here, a concentric nozzle made of two coaxial capillaries (Figure 5a) is used to separate the two solutions and spraying them simultaneously which forms stable core/shell microcapsules. Both the coaxial capillaries are at same potential. The active/core material flows through the central capillary and the wall/shell material flows through the annular nozzle. Advantages associated with coaxial ESPR are effective protection of bioactivity, uniform size distribution, and high encapsulation efficiency (EE) (Bakry et al. 2016; Jaworek and Sobczyk 2008). In coaxial ESPR equipment (Figure 3i), one needle has a larger diameter than other so that the two separated solutions are released concurrently. Here encapsulation of bioactive compound can be done in two ways. In first case, one solution contains a polymer, while the second solution contains the bioactive compound. This case is used when bioactive can be easily electrosprayed. In second case, one solution contains mixture or polymer and bioactive compound, while the second contains the polymer only. This case is used when ESPR cannot be done efficiently due to intrinsic properties of bioactive compound, thus

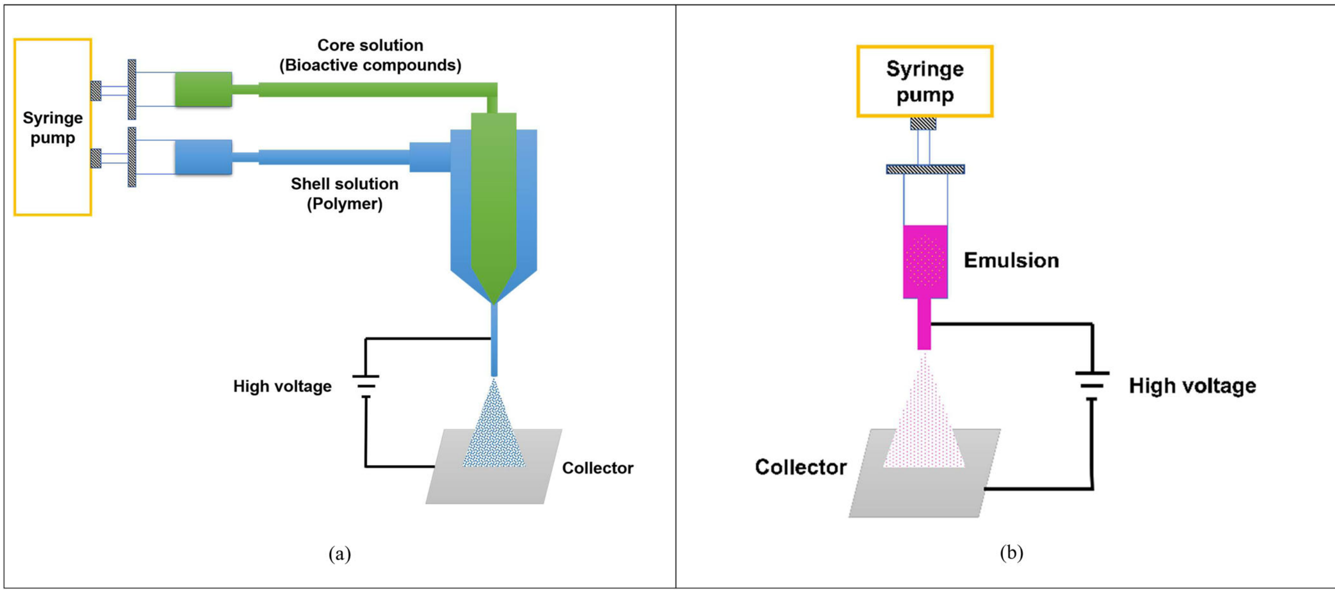

Figure 5. Schematic illustration of (a) co-axial electrospraying and (b) emulsion electrospraying (Adapted from Niu, Shao, Luo 2020).

polymer is used as a carrier. The bioactive compound and the polymer can interact through hydrogen bonds in both cases (Tapia-Hernández, Rodríguez-Félix, and Katouzian 2017). Electro-coextrusion is the term used for EPSR from a co-axial nozzle. Other nozzles like double nozzle (Figure 3j) which is made of two parallel capillaries produces janus particles whereas dual nozzle (Figure 3k) made of two capillaries having a common outlet forms particle/matrix microcapsule. To obtain core/double shell microcapsule three coaxial nozzle (Figure 3l) is used and to obtain three color janus particles triple nozzle (Figure 3m) of three parallel capillaries is used (Jaworek, Sobczyk, and Krupa 2018).

In order to prepare a strong multicore shell composite matrix for water insoluble bioactives, emulsion ESPR (Figure 5b) is an effective technique. Here, bioactive compounds are initially made to entrap inside the droplets of emulsion (formed using homogenizer and emulsifier), and then adding shell polymer in it to make ESPR solution. This solution is then electrosprayed, during which the shell polymer is solidified and it coats the emulsion in order to obtain micro/nano particles having a core-shell matrix. Instead of physical separation as in case of coaxial ESPR, formation of particles is due to solidification and coating of polymer on emulsion. Properties of emulsion (droplet size, viscosity, emulsion stability, etc.) are important in preparation of particles (Niu et al. 2020b). Proteins are especially interesting matrices for emulsion ESPR because of their amphiphilic structures, which enables them to act as effective emulsifiers (Gómez-Mascaraque et al. 2017b).

Applications in food

Electrosprayed particles have functional and structural advantage which is why they have been used for chocolate processing, film coating, stabilization of food ingredients, and encapsulating nutraceuticals (Raval et al. 2019). Bioactives like polyunsaturated lipids, alkaloids, probiotics, carotenoids, phenolics, peptides, and organosulfur compounds have gotten a lot of recognition due to their disease preventing and health enhancement bio-functionalities. The bulk of these compounds are extremely unstable and easily oxidized during storage and processing, particularly when exposed to light or oxygen and/or high temperature. Encapsulating them gives an advantage of controlled release and improves their bioavailability along with preserving them (Niu et al. 2020b). For encapsulation of these bioactive substances, ESPR has been extensively explored. In particular, zein and gelatin have demonstrated excellent encapsulation profiles and stabilities by containing β-Carotene (Gómez-Mascaraque et al. 2017b), epigallocatechin gallate (EGCG) (Gómez-Mascaraque et al. 2019), green tea extract (catechins) (Gómez-Mascaraque et al. 2017a), saffron extract (Alehosseini et al. 2019a), chanar extract (Costamagna et al. 2017), curcumin (Alehosseini et al. 2019b), and resveratrol (Jayan et al. 2019). Encapsulation of these compounds using ESPR had good EE with particle size upto micron and nano scale, provided good stability to core material and retained the properties of active compound which widens its spectrum and thus widening its scope for food applications (Castro Coelho, Nogueiro Estevinho, and Rocha 2021). Encapsulation provided protection against oxidation, released fewer off flavors, maintaining characteristics properties, better thermal stability, and controlled release (Table 2).

Various authors have worked on microencapsulating probiotics like *L. plantarum* and *B. lactis* (Coghetto et al. 2016; Gómez-Mascaraque et al. 2017c; Zaeim et al. 2019) using ESPR. In a study by Gómez-Mascaraque et al. (2017c), coaxial ESPR was used to microencapsulate probiotic bacteria *L. plantarum* with WPC cores and outer layer of gelatin. Gelatin solution was prepared in acetic acid. In comparison with uniaxial ESPR it was found that loss in *L. plantarum* colony formation unit increased during coaxial ESPR. Bacterial count reduced rapidly in gelatin-coated probiotics during storage in freezer. Also, they were unable to protect the encapsulated material from humid environment, thermal stresses, or during digestions. Low pH and osmotic

Table 2. Application of ESPR for encapsulating food and food derived products.

S. no.	Encapsulated material/ coating material	Working conditions	Major outcomes/particle size after treatment	References
1.	β-carotene/WPC	• V:- 14 kV • Flow rate:- 0.3 mL/h • Tip to collector distance:- 7 cm	• EE:- 90% • Capsule size up to <100 nm • Encapsulation successfully stabilized β-carotene against photo-oxidation	López-Rubio and Lagaron (2012)
2.	DHA/zein prolamine	• V:- 12 kV • Flow rate:- 0.2 mL/h • Tip to collector distance:- 15 cm	• Stability of DHA was improved • Better protection against oxidation • Released less off-flavors	Torres-Giner et al. (2010)
3.	EGCG and linolenic acid/zein and gelatin	• V:- 13-18 kV • Flow rate:- 0.5 mL/h	• EE:- above 60% • Enhanced protection of bioactive	Gómez-Mascaraque et al. (2019)
4.	EGCG/gelatin	• V:- 15–28 • Flow rate:- 0.15–0.5 mL/h • Tip to collector distance:- 10	• EE:- 100% • Retained antioxidant activity • Prevented degradation	Gómez-Mascaraque, Lagarón, and López-Rubio (2015)
5.	Folic acid/WPC	• V:- 10 kV • Flow rate:- 0.15 mL/h • Tip to collector distance:- 9–11 cm	• ESPR produced smaller capsules with better particle size distribution than nano-spraying • Folic acid stability was improved	Pérez-Masiá et al. (2015)
6.	Maltol/stearic acid and ethyl cellulose	• V:- 13–15 • Flow rate:- 10 and 15 µL/min	• 10–100 nm • EE:- 69%	Eltayeb et al. (2013)
7.	Oregano-olive oil/PVA and chitosan	• V:- 19–20 kV • Flow rate:- 0.4–0.6 mL/h • Tip to collector distance:- 9–13 cm	• Antimicrobial effect of oil increased • Antioxidant activity of oil was well maintained	Vehapi, Yilmaz, and Özçimen (2020)
8.	Resveratrol/zein	• V:- 14 kV • Flow rate:- 0.5 mL/h	• Controlled release and better stability of resveratrol • Encapsulating resveratrol in zein protected it from high acidic conditions in stomach	Jayan et al. (2019)
9.	Chanar extract/zein	• V:- 13 kV • Flow rate:- 0.15 mL/h • Tip to collector distance:- 10 cm	• Encapsulation increased stability • Effectively protected bioactive polyphenols and did not affect biological activity of extract	Costamagna et al. (2017)
10.	Saffron extract/zein	• V:- 15 kV • Flow rate:- 0.15–0.20 mL/h • Tip to collector distance:- 10 cm	• Very high EE up to 97% • Increase in stability of extracts at different pH, temperature and UV exposure • Complete release of extract on dissolution	Alehosseini, Gómez-Mascaraque, Ghorani, et al. (2019)
11.	Green tea extract/zein and gelatin	• V:- 13–15 kV • Flow rate:- 0.15–0.20 mL/h • Tip to collector distance:- 10 cm	• Particle size up to 0.5 µm • Better thermal stability	Gómez-Mascaraque (2017a)
12.	*Chlorella protothecoide/* sodium alginate and chitosan biopolymers	• V:- • Flow rate:- 0.5 mL/h	• 123–610 nm • EE:- 77%	Karakaş and Özçimen (2020)
13.	*Origanum vulgare* essential oil/chitosan	• V:- 16 kV • Flow rate:- 0.2 mL/h	• 290–483 nm • EE:- 70.1–79.6% • Improved antifungal property	Yilmaz et al. (2019)
14.	L-5-methyltetrahydrofolate/ pullulan and glucose syrup	• V:- 16–20 kV • Flow rate:- 0.001–0.007 mL/h • Tip to collector distance:- 15 cm	• For 21 d storage encapsulated material showed higher stability and recovery than free	Svarc et al. (2020)
15.	Phycocyanin/PVA	• V:- 15–25 kV	• 395 ± 71 nm • EE:- 75% • Thermal resistance up to 216 °C • Preserved antioxidant activity	Schmatz et al. (2020)
16.	Quercetin/zein	• V:- 15 kV • Flow rate:- 0.1 mL/h • Tip to collector distance:- 15 cm	• EE:- 87–93% • Spherical morphology • Greater bioavailability and stability	Rodríguez-Félix et al. (2019)
17.	*Coccoloba uvifera* L./ agave fructans	• V:- 0–30 kV • Flow rate:- 0.1 mL/h • Tip to collector distance:- 22 cm	• 655–7250 nm, spherical particles • Controlled release of encapsulated extract	Ramos-Hernández et al. (2021)
18.	Broccoli extract/zein	• V:- 16 kV • Flow rate:- 1 mL/h • Tip to collector distance:- 10 cm	• Uniform morphology capsules • High thermal stability	Radünz et al. (2020)
19.	Broccoli extract/zein	• V:- 16 kV	• EE:- 97.8%	Radünz et al. (2021)

(continued)

Table 2. Continued.

S. no.	Encapsulated material/ coating material	Working conditions	Major outcomes/particle size after treatment	References
		• Flow rate:- 1 mL/h • Tip to collector distance:- 10 cm	• Controlled oxidation processes and hyperglycemia	
20.	Lycopene/WPC	• V:- 12–18 kV • Flow rate:- 0.15 mL/h • Tip to collector distance:- 9–20 cm	• EE:- 75% • Good protection against heat and moisture	Pérez-Masiá, Lagaron, Lopez-Rubio (2015)
21.	Lycopene/zein	• V:- 14 kV • Flow rate:- 0.3–1 mL/h • Tip to collector distance:- 10 cm	• 0.36 μm • Protection against degradation and stability against light	Kose and Bayraktar (2016)
22.	Labdanum essential oil bioactive/β cyclodextrin	• V:- 8–22 kV • Flow rate:- 0.12–0.35 mL/h	• 25–160 nm • Successful entrapment of volatile bioactive present in labdanum essential oil	Kose, Tekin, and Bayraktar (2021)
23.	Ursolic acid/gelatin	• V:- 11–14 kV • Flow rate:- 0.15 mL/h • Tip to collector distance:- 15 cm	• 706–752 nm • Crystalline nature of ursolic acid changed to amorphous • Stability up to 90 °C • Controlled release and improved bioaccesibility	Karimi et al. (2020)
24.	*Chlorella protothecoides* oil/ chitosan and sodium alginate	• V:- 12 kV • Tip to collector distance:- 15 cm	• 129–610 nm • EE:- 60–77%	Karakaş and Özçimen (2020)
25.	Aspalathin/Eudragit S100	• V:- 20 kV • Tip to collector distance:- 15 cm	• <1.1 μm • EE:- 50% • Better encapsulation than conventional methods • Controlled release	Human et al. (2019)
26.	Curcumin/walnut protein isolate	• V:- 7.5–22.5 kV • Flow rate:- 0.06–0.2 mL/h • Tip to collector distance:- 10–30 cm	• 104–180 nm • EE:- 61% • Controlled release	Asadi et al. (2021)
27.	Fish oil/Kafirin	• V:- 20–25 kV • Flow rate:- 0.5–1 mL/h	• 552–861 nm • EE:- 95%	Cetinkaya et al. (2021)
28.	Sardine oil/gliadin proteins	• V:- 15–20 kV • Tip to collector distance:- 10–20 cm	• 183–957 nm • Increase in oxidative stability	Dórame-Miranda et al. (2021)
29.	*Lactobacillus plantarum*/ sodium alginate citric pectin matrix	• V:- 24 kV • Flow rate:- 2 mL/h • Tip to collector distance:- 15 cm	• 7–2000 μm • Protection against gastric juices and controlled release • Improved storage stability	Coghetto et al. (2016)
30.	*Lactobacillus plantarum*/WPC and gelatin	• Flow rate:- 0.15 mL/h • Tip to collector distance:- 10 cm	• 0.6 μm • Unable to protect the active compound • Type of solvent is important for better encapsulation results	Gómez-Mascaraque (2017c)
31.	*Lactobacillus plantarum*/WPC	• V:- 10–14 kV • Flow rate:- 0.15 mL/h	• High productivity and very low loss of active compound • Similar protection provided against digestion as by freeze drying	Gómez-Mascaraque et al. (2016)
32.	Lactobacillus plantarum and Bifidobacterium lactis/ inulin and resistant starch	• V:- 9.5 kV • Flow rate:- 5 mL/h • Tip to collector distance:- 100 mm	• Micron size particles containing prebiotics and probiotics produced • Viability and survival of probiotics improved	Zaeim et al. (2019)
33.	Glucose oxidase/ chia mucilage	• V:- 17 kV • Flow rate:- 2 mL/h • Tip to collector distance:- 5.5 cm	• EE:- 92% • Protecting capsule against breakdown • Enzyme denaturation at 94 °C • For slowing down release jet mode is effective and to increase release rate dripping mode is effective	Rentería-Ortega et al. (2021)

shock created by acetic acid was attributed to badly impacting on cell viability. Thus, type of solvent used is very important for the encapsulated material. Similarly, Zaeim et al. (2019) used ESPR for probiotics and prebiotics double layer co-encapsulation. In Ca-alginate/chitosan ESPR microcapsules, Zaeim et al. (2019) co-encapsulated *L. plantarum* and *B. lactis* with either inulin or resistant starch. Adding resistant starch to *B. lactis* microcapsules improved bacterial survival considerably. In general, resistant starch microcapsules demonstrated a better performance in gastrointestinal conditions than those that contain inulin to preserve the viability of the probiotics. However, microcapsules that contain inulin enhance lactobacilli survival more effectively than those containing starch during storage. Coghetto et al. (2016) encapsulated *L. plantarum* in a sodium alginate citric pectin matrix and it was found that ESPR resulted in formation of complex between pectin and alginate which resulted in protecting the active compound from intestinal and gastric juices; and controlled survival of microencapsulated cells in refrigeration conditions. In another study, *L. plantarum* was encapsulated within WPC matrix. It was reported that the encapsulation of fresh culture in place of freeze-dried bacteria led to lowering viability loss during processing and had greater productivity. When compared with freeze dried preservation method, ESPR particles provided similar protection against digestion (Gómez-Mascaraque et al. 2016). These findings showed that ESPR is an effective technique to encapsulate these probiotics and increases their applications in food formulations.

Vehapi, Yilmaz, and Özçimen (2020) used ESPR to enhance bioactive compound's antimicrobial activity in essential oils. They encapsulated a mixture of olive oil and oregano essential oil by chitosan polymer. EE of 90.78% and 89.56% was reported when nanoparticles were loaded with 0.5–2% and 0.1–2% oregano-olive oil, respectively. These high EE values indicate ESPR as a highly efficient encapsulation process for essential oils. Further, the antifungal effectiveness of oregano-olive oil against *Colletotrichum gloeosporioides, Botrytis cinerea, and Penicillium chrysogenum* was improved by encapsulating with chitosan. According to the findings of this study, oil-loaded nanoparticles have the potential to be used as an antimicrobial agent in food preservation. *Chlorella protothecoides* oil encapsulated inside chitosan and sodium alginate using EPSR formed particles upto nano level. Particle size decreased as the concentration of polymer used and the distance between tip and collector increased. It was also discovered that as the flow rate increased, the particle size increased (Karakaş and Özçimen 2020). Encapsulating sardine oil and using pecan nutshell extract and gliadin proteins along with it increased its oxidative stability. Due to antioxidant properties of pecan nut shell and modification in structures brought by interaction between gliadin and sardine oil, oxidative stability increased (Dórame-Miranda et al. 2021). *Origanum vulgare* essential oil loaded in chitosan nanoparticles using EPSR provided good EE along with providing good preservation and controlling their release. In addition to this essential oil's antifungal effect against *Alternaria alternata* AY1 improved by encapsulating it in chitosan as both have antifungal activity present individually (Yilmaz et al. 2019). In another study, inclusion complexes were formed using electric field for selectively isolated volatile bioactive compounds present in labdanum essential oil. Successful entrapment of active compound was attributed to β-cyclodextrin/volatile inclusion complexes formed due to hydrophobic interactions, van der waals force and hydrogen bonds (Kose, Tekin, and Bayraktar 2021). Another research used ESPR to encapsulate fish oil inside kafirin (hydrophobic in nature) with EE up to 95% (Cetinkaya et al. 2021).

ESPR was also beneficial in nanoencapsulating aspalathin (hydrophilic dihydrochalcone bioactive) which has poor bioavailability and stability, restricting its use as functional food ingredient. Using synthetic polymer as a wall material, EE of upto 50% was achieved and had high loading capacity as compared to particles produced using conventional methods (centrifugation) (Human et al. 2019). Quercetin-loaded zein particles using ESPR had an EE ranging from 87% to 93%. In this study, it was observed that trapped quercetin had 20% less in vitro gastrointestinal release and 4% more in vitro bioavailability than free quercetin. Interactions found between quercetin and zein were hydrogen bonds. In TEM images, it was found that quercetin was entrapped inside zein and was distributed throughout; and had compact and spherical morphology (Rodríguez-Félix et al. 2019). In encapsulation of ursolic acid inside gelatin, it was found that mean diameter increased and morphology was dissimilar to electrosprayed gelatin particles alone which had a smooth morphology. This change was due to development of hydrophobic interactions and hydrogen bonds amid gelatin and ursolic acid which might have altered structure of proteins. Similarly, addition of sodium dodecyl sulfate improved morphology and uniformity of particles which was attributed to reduction in surface tension which would have further decreased the droplets Rayleigh limit resulting in formation of small particles by phenomenon known as coulomb fission (Karimi et al. 2020). Phenolic compounds and glucosinolates present in broccoli are affective in killing tumor cells, but these compounds have very low stability. Encapsulation of these compounds using ESPR was done by Radünz et al. (2020), and it was found that formed particles had uniform morphology along with good thermal stability. These encapsulated extracts showed selective cell death against glial tumor cells without being toxic to non-tumor cells. In another study by Radünz et al. (2021), it was reported that encapsulating broccoli extracts inside zein showed spherical morphology and high EE. Encapsulation promoted control of hyperglycemia and oxidative stresses. For encapsulation of EGCG using whey protein isolate (WPI) and bacterial cellulose, emulsion ESPR was used. Promising results were shown by this technique as it produced emulsion with smaller particle size and lower viscosity with good storage stability and protected active compound from heat, moisture, and dissolution conditions (Paximada et al. 2017). EGCG was also encapsulated by Gómez-Mascaraque, Lagarón, and López-Rubio (2015) using ESPR and it was found that using gelatin as a wall material 100% EE was achieved along with retained the antioxidant activity of EGCG. Gelatin prevented degradation of core material in aqueous solution (pH $=$ 7.4) which might have happened due to stabilization brought by intermolecular interactions between the core and wall material. After in-vitro digestion, the coaxial ESPR method used to encapsulate EGCG within zein and gelatin retained its antioxidant properties and protected it from thermal degradation (Gómez-Mascaraque et al. 2019). L-5-methyltertahydrofolate, a naturally occurring form of folate has less stability than synthetic folate which limits its application for food fortification. In a study by Svarc et al. (2020), combination of pullulan and glucose sirup were used to encapsulate L-5-methyltertahydrofolate in order to provide it a higher stability. Reports showed that electrosprayed capsules had excellent stability when stored for 21 d whereas free L-5-methyltertahydrofolate significantly decreased by 40%. Micronizing curcumin using ESPR helped it improve its aqueous solubility and thus increasing its bioavailability. Increase in solubility was attributed to hydrophilic PVP used along curcumin along with increase in surface area due to reduction in particle size (Chhouk et al. 2018). In another experiment walnut protein isolate was EPSR to produce nanoparticles in order to entrap curcumin inside it. Curcumin was successfully entrapped with EE of upto 61%. In vitro digestion studies revealed that encapsulated curcumin showed limited release in stomach whereas in small intestine the walnut protein isolate hydrolyzed and curcumin released (Asadi et al. 2021). WPC microparticles can be obtained over a wide pH range. β-carotene was encapsulated inside WPC and it was found that it successfully protected it against photooxidation (López-Rubio and Lagaron 2012). The emulsion ESPR encapsulation of β-carotene within zein and WPC enhanced its bioaccessibility following in-vitro digestion (Gómez-Mascaraque et al. 2017b). In a research carried out by Basar et al. (2020), deep eutectic solvent (DES) (mixture of choline chloride and butanediol) was used as green alternative for encapsulating β-carotene. These solvents have high solubilization power, high stabilization, and extraction ability along with a wide range of polarity and can exist in liquid state far below 0 °C. Smooth and spherical β-carotene encapsulated particles in WPC were obtained using DES formulation and ESPR with a high loading capacity. It was also observed that β-carotene without any encapsulation oxidized completely under UV light in 3 h, where WPC coated β-carotene oxidized only about 20% (Basar et al. 2020). In another study β-carotene was encapsulated inside pullulan-WPI system which increased its stability and protected against heat and humidity. Introduction of WPI and 3% pullulan with 1:50 (core:wall) for microencapsulating β-carotene yielded a compact structure and particles had round shape. With lowering relative humidity and temperature, the preservation of β-carotene from electrosprayed particles increased dramatically (Niu et al. 2020a).

Emulsion ESPR (Table 3) was compared with spray drying for encapsulating thermosensitive α-linolenic acid by Gómez-Mascaraque and López-Rubio (2016) in which WPC and SPI were used as emulsion stabilizers and wall material; and it was observed that spray drying resulted in destruction of bioactive whereas ESPR was a good alternative in preserving these bioactive by achieving good EE. At 80 °C, accelerated degradation assays were performed and encapsulated particles using ESPR delayed degradation. It was also found that emulsions prepared using sonication had less EE due to the heat generated by it and using gelatin for emulsions along with acetic acid as a solvent degraded bioactive due to low pH (Gómez-Mascaraque and López-Rubio 2016). D-limonene was nanoencapsulated within *Alyssum homolocarpum* seed gum using emulsion ESPR. Emulsion was formed using high-pressure homogenizer or ultrasonic homogenizer and Tween 20. Compact, smooth, and round nanocapsules were obtained which had good storage stability (Khoshakhlagh et al. 2018). In a study by Pérez-Masiá, Lagaron, Lopez-Rubio (2015), spray drying and emulsion ESPR were, respectively, used in order to encapsulate lycopene using three different edible biopolymer matrices that are WPC, chitosan, and dextran. Spray drying showed very poor EE along with affecting lycopene's stability due to high temperature used in it. Whereas ESPR especially using WPC as a wall material showed excellent EE upto 75% along with better protection against thermal degradation and moisture. Similarly, Kose and Bayraktar (2016) encapsulated lycopene inside zein using ESPR and found that increasing the concentration of zein in solution mixture produced more smoother and spherical structures. Encapsulation protected lycopene from degradation and provided stability against light. From these studies, it can be concluded that emulsion ESPR is far better than spray drying in terms of providing excellent EE and providing stability to active compound by processing them non-thermally.

To minimize agave fructans capsules hygroscopicity from 12% to 8%, whey protein has been added as a stabilizer. ESPR was used to encapsulate extracts of sea grape leaf in this matrix. Spherical particles in nano range were obtained which had an improved thermal stability. The matrix was also successful in protecting the extracts until it reaches the small intestine where it was completely released thus increasing its biological activity and bioavailability (Ramos-Hernández et al. 2021). Using ESPR, phycocyanin was encapsulated inside polyvinyl alcohol (PVA) and ultrafine particles were obtained. Encapsulation increased thermal resistance of phycocyanin up to 216 °C and retained its antioxidant property, as measured by DPPH and ABTS activity (Schmatz et al. 2020). ESPR was also used to form Phycocyanin/PVA based film which had good mechanical properties, thermal resistance, and antioxidant activity (Schmatz, Costa, and Morais 2019).

Atomization of chocolate was improved using ESPR, by reduction in particle size provided by it and increasing the overall area covered by it after spraying. It was also observed that particle size reduced on reducing the yield value, electrical resistivity, and viscosity of chocolate which were manipulated by varying the amount of lecithin, temperature, and fat content with improved spraying quality (Gorty and Barringer 2011). Glucose oxidase used widely in bakery industry is highly prone to loss due to processing. Encapsulation done by Rentería-Ortega et al. (2021) inside chia mucilage protected it from thermal, pH, and mechanical stresses along with slowing its release rate which was attributed to structural change brought by ESPR. In a study by Sreekumar et al. (2017), it was reported that chitosan solutions having low to medium conductivity values were optimum for ESPR. Conductivity values of chitosan were dependent on its physicochemical properties and degree of acetylation. Upto 10% degree of acetylation in chitosan showed reduced solution conductivity. In a study by

Table 3. Application of EAPG and emulsion ESPR for encapsulating food and food derived products.

S. no.	Techniques	Encapsulated material/ coating material	Working conditions	Major outcomes/particle size after treatment	References
1.	EAPG	DHA enriched fish oil/zein	• V:- 20 kV • Flow rate:- 10 mL/min • Air pressure:- 10 L/min	• 1.4 μm • EE:- 84 % • Did not affect sensory properties of food product on fortification	Busolo et al. (2019)
2.	EAPG	DHA rich algae oil/WPC	• V:- 0–30 kV • Flow rate:- 1 mL/min • Air pressure:- 10 L/min	• 5 μm • Good oxidative stability • No impact on sensory properties of fortified food	Prieto and Lagaron (2020)
3.	EAPG	Fish oil/WPC, pullulan and glucose sirup	• V:- 10–15 kV • Flow rate:- 1.5–1.8 mL/min	• <3 μm • EE:- 85% • Glucose sirup provided better oxidative stability than dextran	García-Moreno et al. (2018)
4.	EAPG	Fish oil/zein	• V:- 10 kV • Flow rate:- 1.4 mL/min • Air pressure:- 10L/min	• 2–3 μm • EE:- 83% • Improved oxidative stability of fortified mayonnaise	Miguel et al. (2019)
5.	EAPG	EPA-rich oil/WPC	• V:- 10 kV • Flow rate:- 30 mL/h • Air pressure:- 10 L/min	• 5 μm • EE:- 80% • Increased oxidative and thermal stability	Escobar-García et al. (2021)
6.	Emulsion ESPR	*β*-carotene/WPC	• V:- 17–23 kV • Flow rate:- 50–100 μL/h • Tip to collector distance:- 15 cm	• 2.51 μm • Provided protection against UV light	Basar et al. (2020)
7.	Emulsion ESPR	*β*-carotene/zein and WPC	• V:- 13–16 kV • Flow rate:- 0.15 mL/h • Tip to collector distance:- 10 cm	• High EE obtained from zein emulsion • Improved bioaccessibility of active compound after in vitro digestion	Gómez-Mascaraque (2017b)
8.	Emulsion ESPR	*β*-carotene/pullulan-WPI	• V:- 18 kV • Flow rate:- 0.01 mL/h • Tip to collector distance:- 15 cm	• Good stability and protection against heat and moisture	Niu, Shao, Feng (2020)
9.	Emulsion ESPR	*α*-linolenic acid/WPC	• V:- 15 kV • Flow rate:- 0.15 mL/h • Tip to collector distance:- 10 cm	• EE:- 62–72% • Delayed degradation at 80 °C	Gómez-Mascaraque and López-Rubio (2016)
10.	Emulsion ESPR	D-limonene/*Alyssum homolocarpum* seed gum	• V:- 15 or 20 kV • Flow rate:- 0.05 or 0.1 mL/h • Tip to collector distance:- 15 cm	• EE:- 73.4% • Improved storage stability • Smooth and compact nanocapsules	Khoshakhlagh et al. (2018)
11.	Emulsion ESPR	EGCG/WPI	• Tip to collector distance:- 10 cm	• EE:- 97% • Protection against heat, light and dissolution conditions	Paximada, Kanavou, and Mandala (2020)

Paximada, Kanavou, and Mandala (2020), suspensions containing bacterial cellulose and WPI were electrosprayed and it was found that electrical conductivity, viscosity, and surface tension increased with increase in concentration of bacterial cellulose which can be attributed to change in bonds formed between proteins and polysaccharides. The particle size of product formed was at nano level and it was concluded that structure of these electrosprayed particles could be predicted by interfacial viscosity and surface tension (Paximada, Kanavou, and Mandala 2020).

Electrospray coating (Table 4) is a modern process for generating small droplets from conductive or semi-conductive feeding solutions down to submicron sizes. This approach enables homogeneous surface coatings to be developed using a limited quantity of coating materials (Cakmak, Kumcuoglu, and Tavman 2019). ESPR has been utilized widely for coating food surfaces with lipid-based coatings like butter (Khan et al. 2013c), sunflower oil (Khan et al. 2013b, 2013c) and chocolate (Khan et al. 2014; Luo et al. 2012) based coatings. Khan et al. (2013a) coated apple slices using chocolate based or water in oil (w/o) emulsion-based coating. Coating by olive oil-based w/o emulsion was better in terms of anti-browning properties whereas chocolate-based coating was better in terms of reducing moisture loss. W/o-based emulsion penetrates the surface of the apple thus the film formed on the surface was thin which wasn't effective in reducing water loss whereas chocolate-based film does not penetrate. Similarly, Cakmak, Kumcuoglu, and

Table 4. Application of ESPR for micronizing, coating, and film formation for food and food derived products.

S. no.	Purpose	Encapsulated material/ coating material	Working conditions	Major outcomes/particle size after treatment	References
1.	Coating	Polyglycerol polyricinoleate based w/o emulsion (Sunflower oil)	• Flow rate:- 15 mL/h • V:- 9–18 kV	• 40–60 μm • Addition of whey protein isolate enhanced water vapor barrier properties of the film formed	Khan et al. (2013b)
2.	Coating	Apple slices/ w/o (olive oil)	• Flow rate:- 15 μL/min • Tip to collector distance:- 15 cm • V:- 10–13 kV • Time:- 25 s	• ESPR had better water vapor barrier properties, coating homogeneity and utilized less coating material than dip coating	Cakmak, Kumcuoglu, and Tavman (2018)
3.	Coating	Apple slices/chocolate based and w/o emulsion-based coating	• V:- 20–25 kV	• Electrospraying w/o emulsion on slices showed anti browning properties • Moisture loss was reduced more by chocolate-based coating than w/o emulsion	Khan et al. (2013a)
4.	Micronization	Curcumin/ polyvinylpyrrolidone	• V:- 10 kV • Tip to collector distance:- 15 cm	• 160–730 nm • Micronization by ESPR improved aqueous solubility of curcumin	Chhouk et al. (2018)
5.	Micronization	Hydroxypropyl methyl cellulose (HPMC)	• V:- 10–18 kV • Flow rate:- 0.5–1 mL/h • Tip to collector distance:- 15–20 cm	• 79–161 nm HPMC high molecular weight • 833–1188 nm HPMC low molecular weight • Concentration was an important parameter for particle morphology	Silva et al. (2021)
6.	Packaging film production	PVA containing chitosan	• Flow rate:- 10 μL/min • V:- 15 kV	• Film produced had better mechanical properties, lower oxygen permeability, higher water barrier properties and antibacterial activity than control	Liu, Wang, and Lan (2018)
7.	Film formation	Zein	• V:- 7.8–8.7 kV • Tip to collector distance:- 15 cm	• Zein film was produces successfully without affecting water vapor barrier capacity	Gaona-Sanchez et al. (2015)
8.	Coating	Sunflower oil and chocolate-based coating	• V:- 20-25 kV • Flow rate:- 0.9 mL/h	• ESPR sunflower oil resulting in smaller droplets and homogeneous distribution than chocolate-based coating • Chocolate based coating had better barrier efficiency	Khan et al. (2014)
9.	Coating	Strawberries/w/o emulsion	• V:- 10–13 kV • Tip to collector distance:- 14 cm • Flow rate:- 3.5 mL/h	• Coating by ESPR reduced moisture loss and maintained its firmness during storage • Sensory properties were not affected	Cakmak, Kumcuoglu, and Tavman (2019)
10.	Coating	Strawberry/alginate based coating enriched with methyl cinnamate and carvacrol	• V:- 85 kV	• ESPR coated strawberries had better evenness and coating efficiency than conventional methods • Better in color retention, fruit firmness, low fruit weight reduction, delaying decay and microbial spoilage than conventional methods	Peretto et al. (2017)
11.	Packaging film production	Phycocyanin-PVA/ polycaprolactone and poly-L-lactic acid	• Tip to collector distance:- 180 mm • V:- 20 kV • Flow rate:- 50 μL/h	• EE:- 75% • Film formed had good thermal resistance, mechanical properties and good antioxidant activity	Schmatz, Costa, and Morais (2019)
12.	Bilayer film formation	Zein and Pectin bilayer	• V:- 12.5 kV and Flow rate:- 4 mL/h (pectin film) • V:- 8.3 kV and flow rate:- 14.5 mL/h	• Up to 100 • No chemical reaction between two layers	Gaona-Sanchez et al. (2021)

Tavman (2018) coated apple slices by ESPR w/o emulsion on it. It was found that coating by using ESPR had better homogeneity, water vapor barrier properties, showed less color change while storage and utilized less coating material than dip coating method. Sunflower oil-based coating had smaller droplets and homogeneous distribution than chocolate-based coating. Chocolate-based coatings were thicker and had limited "wrap around effect" but exhibited better barrier properties than sunflower oil-based coating (Khan et al. 2014). Addition of WPI by Khan et al. (2013b) to polyglycerol polyricinoleate based emulsions film coating increased its water vapor barrier properties which can be attributed to network formation by proteins leading to increase in mechanical stability of film or prevention of cracks. ESPR alginate-based coating enriched with methyl cinnamate and carvacrol on strawberries delayed decay and microbial spoilage, low moisture loss and was better in maintaining fruit firmness than the conventional methods of coating (Peretto et al. 2017). Similarly, Cakmak, Kumcuoglu, and Tavman (2019) coated strawberries using w/o emulsion and found that coating using ESPR reduced moisture loss and maintained product's firmness and sensory properties. A flexible biodegradable yellow colored bilayer film was successfully produced from zein (hydrophobic) and pectin (hydrophilic) using ESPR by Gaona-Sanchez et al. (2021) with particle size upto 100 µm. There was no chemical reaction between the two layers and the surface formed had a good continuous structure along with well-defined interface. This opens an opportunity to create an intelligent bioactive film in an environmentally friendly manner. In a study mechanical strength of paperboard packaging was improved by coating it with kaolin/calcium carbonate and electrosprayed polylactic acid along with improving its water vapor transmission rate (Thitsartarn and Jinkarn 2021).

ESPR showed excellent coating and EE, micronization potential, reduced quantity of coating solution required and formed a homogeneous film on irregular food surfaces which can help in reducing industrial manufacturing costs. Wall material properties, solvent properties, core material properties, process parameters, and environmental conditions are important in order to obtain optimum encapsulation, coating, and overall process efficiency. Still applying the above studies on industrial scale is challenge as ESPR has very low throughput. ESPR setup which can provide a good throughput has been discussed in further sections.

Advantages and disadvantages of ESPR on conventional techniques

ESPR is a non-thermal treatment using low energy, low cost, and is a rapid single step processing method with high versatility. ESPR technique develops stable micron size particles with droplet size <1 µm which is smaller than conventional mechanical atomizers and particles formed have uniform size distribution. EPSR protects bioactive compounds through high EE and allows for regulated and sustained release (Bhushani and Anandharamakrishnan 2014; Bakry et al. 2016; Jaworek and Sobczyk 2008; Khan, Nazir, and Maan 2017). It is widely and very effectively applicable to hydrophobic and water insoluble bioactive compounds (Rostamabadi et al. 2021). Since it does not require extreme temperatures, pressures, or harsh chemicals, ESPR is a promising tool for the encapsulation of bioactive compounds (Niu et al. 2020b). It can be carried out under ambient conditions of temperature and pressure. In this method, denaturation of enzymes and proteins is reduced, that is due to very limited exposure and rapid processing. In case of coating with ESPR moisture permeation is reduced and the amount of coating material required than dip coating is also less. Coating by using ESPR droplets deposits on all sides of target material that is due to the wrap around effect (Bhushani and Anandharamakrishnan, 2014; Bakry et al. 2016; Jaworek and Sobczyk 2008; Khan, Nazir, and Maan 2017). ESPR provides an advantage of coating at higher growth rate with similar coating morphology throughout (Jaworek, Sobczyk, and Krupa 2018). Other advantages are nanoencapsulation in one step is done here without further drying. Particle formation upto nano range has the added benefit of reducing their effects on the textural properties of food items. Ability to construct multilayer encapsulated structure in one step, with no need of additional coating by using coaxial or multilayer configurations is another benefit. It is easy and inexpensive to run if the process conditions are optimized, and consistency checks on the particles can be performed easily by interrupting the process for a few seconds (Gómez-Mascaraque and Lopez-Rubio 2019). In comparison to ESPR conventional techniques require several steps, use several different solvents, are costly, generate more residue and do require high energy (Bakry et al. 2016; Baldino, Cardea, and Reverchon 2019b; Tapia-Hernández et al. 2015).

ESPR's disadvantages are that its performance is significantly less than traditional techniques and that implementing it on a broad scale requires more research (Tapia-Hernández et al. 2015). Use of multi needle arrangement or setups such as supercritical assisted ESPR (Baldino, Cardea, and Reverchon 2019a) and EAPG (Prieto and Lagaron 2020) overcomes these limitations which have been discussed in detail in further sections. Using water as a solvent is a must in most of the cases in order to completely remove toxic solvents from final product. However, water's physical properties are not perfect for ESPR processing. Water evaporates slowly from aqueous polymer suspension or solution as it has comparatively a higher boiling point than other organic solvents and has high surface tension. Thus, high voltages are required to attain stable jets; but excessive voltage may lead to ionization of water causing corona discharge (Gómez-Mascaraque and Lopez-Rubio 2019). These drawbacks can be remedied by altering certain critical features of the set-up, for example by using the multiplex (Figure 3n) style system. In this configuration, there are many nozzles or syringes with various geometrical forms, such as oval, triangular, or hexagonal, to generate complex nanoparticles in larger quantities (Tanhaei et al. 2021). Few natural polymers used for encapsulation in ESPR require solvents like acetic acid and ethanol for dissolution which might pose food safety risk and cause undesirable flavor in the end product (Niu et al. 2020b).

Supercritical assisted electrospraying

SCF technology in food processing has been used for biomass disruption, extraction and encapsulation, microbial and enzymatic inactivation. SCFs have properties between gases and liquids; density and dissolving power similar to liquid, compressibility, and diffusivity similar to gases thus making them an excellent solvent (Dhiman and Prabhakar 2021). Using $scCO_2$ in addition to ESPR improves the process by reducing the surface tension and viscosity largely and allows production of micro and nanoparticles of controlled size and uniform particle size distribution. This process was named as Supercritical assisted ESPR (SA-ESPR). In SA-ESPR (Figure 1b), $scCO_2$ is added to the liquid solution containing active components, which is then electrosprayed at atmospheric pressure (Baldino, Cardea, and Reverchon 2019b; Cardea & Reverchon, 2019). Viscosity and surface tension is reduced due to gaseous properties of $scCO_2$. It was found that using SA-ESPR, operating window, and production rates were improved upto hundred times as compared to traditional ESPR (Baldino, Cardea, and Reverchon 2019b). Also using SCF to assist ESPR can be effective in reducing the probability of losing activity of active compound and less analyte loss, also since SCF forms solvent free products, no co-products and does not pollute the environment thus can reduce the economic burden (Demirdöğen et al., 2018). In addition to the all-other working parameters as mentioned in above sections, pumping pressure of $scCO_2$ is important in SA-ESPR as it promotes mixing with the liquid polymeric solution (Baldino, Cardea, and Reverchon 2019b). Using SA-ESPR, polyvinylpyrrolidone (PVP) stable microparticles upto 0.35 µm with narrow particle size distribution were produced. Solution was loaded and pressurized at 8, 10, 12, 14, and 16 MPa at 35 °C. Applied voltage used throughout the process was 30 kV with the help of a generator. Particles were collected on and aluminum stub and sputter coated with gold. It was found that morphology can be easily controlled by this process. It was determined that the method is more robust and versatile than conventional ESPR (Baldino, Cardea, and Reverchon 2019a). In a study by Guastaferro et al. (2020), quercetin was encapsulated inside PVP using SA-ESPR (applied voltage 30 kV at 35 °C and pressurizing solution at 16 MPa) and micron size particles were formed with an average diameter 0.77–2.81 µm. The technique was able to improve the bioavailability of quercetin upto 30 times as compared to unprocessed powder along with preserving its antioxidant activity upto 90%. Using SCF not only improves the ESPR production rates but also opens channels for "green ESPR" and thus has huge potential to be used at industrial level. SA-ESPR has not been explored in the food industry, according to our understanding, and therefore has a lot of research potential.

Electrospraying assisted by pressurized gas

EAPG is a patented and modified ESPR novel micro-encapsulation technique developed by Bioincia (n.d.) which overcomes drawback of low yield of EPSR by producing encapsulated active ingredients in large scale while keeping the benefits of ESPR. EAPG can produce particles with sizes varying from 1 to 10 μm, with rates ranging from 1 to 3 kg of dry powder per hour to 10 kg of dry encapsulated bioactives per hour. For applications in functional foods/nutraceuticals (Table 3), EAPG is well suited especially for encapsulating highly thermo-sensitive bioactive ingredients that require protection against deterioration, controlled release, or isolation from other components (Bioincia n.d.). Fluidnatek™ LE500 Capsultek™ pilot-plant contains an injection unit, a drying chamber, and a cyclonic collector (Figure 1c) (Busolo et al. 2019). The polymer solution is atomized here by pneumatic injector with the help of compressed air which nebulizes under high electric field which is the basis of this innovative high throughput ESPR technology. Solvent gets evaporated inside the evaporation chamber at room temperature, and then the encapsulated substance is collected in powder form (Prieto and Lagaron 2020). Apart from other working parameters as mentioned in above sections, flow rate of air used is an additional important parameter in case of EAPG (Busolo et al. 2019; Prieto and Lagaron 2020). In a study by Busolo et al. (2019), EAPG was used to encapsulate DHA-enriched fish oil inside zein which showed good stability during storage. High EE (upto 84%) was achieved with particle size upto 1.4 µm. On fortifying reconstituted milk with Zein/DHA enriched microcapsules, it was found that the sensory properties were same even after 45 d storage. Since zein is expensive and is water insoluble, in another study (Prieto and Lagaron 2020) WPC and maltodextrin were used to encapsulate DHA-rich algae oil. Spherical particles of about 5 µm were obtained after encapsulation. WPC encapsulated particles had strong oxidative stability, while maltodextrin encapsulated particles were vulnerable to secondary oxidation. On using these both particles in fortification of different food products, it was found that WPC encapsulated particles had least impact on organoleptic properties of food products. Fish oil encapsulated inside a mixture of carbohydrate (dextran or glucose syrup and pullulan) and WPC had shown an EE of upto 85%. In this study, it was found that glucose syrup provided better oxidative stability than dextran which was attributed to its lower molecular weight than dextran which might have led to more densely packed structure and thus limiting oxygen permeability (García-Moreno et al. 2018). In another study, fish oil encapsulated inside zein particles by EAPG were used to fortify low fat mayonnaise and it was found that the capsules were well intact inside the matrix of mayonnaise. Using zein (hydrophobic) as a wall material preserved the emulsion by improving its oxidative stability and was better than using hydrophilic encapsulates. Hydrophilic encapsulates gets disintegrated in water part of emulsion which leads the fish oil to be unprotected and prone to oxidation (Miguel et al. 2019). EPA rich oil encapsulated inside WPC using EAPG formed a compact nanostructure without affecting its bioactivity. Also, an increase in thermal and oxidative stability was observed on encapsulation (Escobar-García et al. 2021). From the above studies, it can be concluded that EAPG has a huge potential to encapsulated thermo sensitive compounds, bioactives and nutraceuticals with high throughput as compared to conventional ESPR technology. To our knowledge, only these were the studies conducted using EAPG until now and therefore have an enormous scope on research end.

Conclusion and future scope

ESPR is a single step processing method that non-thermally processes the product. It can be a commercially attractive technique for encapsulation, coating, micronization, and food packaging. It provided structural advantages like producing small and stable uniform size particles upto nano level, whose size was controlled by manipulating processing parameters like solution type, concentration, applied voltage, flow rate, etc. Process parameters and polymer solution properties had huge influence on morphology. ESPR had shown high encapsulation and coating efficiency. It can protect oxidation-sensitive and heat-sensitive ingredients. ESPR provides better barrier properties than conventional coating techniques due to wrap around effect. Electrosprayed coating had better water vapor barrier properties than conventional techniques; it reduced moisture loss, maintained fruit firmness, low fruit weight reduction, delayed decay, and microbial spoilage. Films formed using ESPR had better thermal resistance, mechanical strength, lower oxygen permeability, and high-water barrier properties. Encapsulation using ESPR protected food micro and macromolecules from photooxidation, acidic conditions, different pH conditions, UV exposure, moisture, thermal stress, mechanical stress, produced less off flavor, and controlled their release without affecting their biological, antibacterial, antifungal, and antioxidant activity, along with improving their bioavailability and bioaccessibility thus forming stable products. Conventional techniques are costly as compared to ESPR and require several steps, generate more residues, use several different solvents, and do require high energy. ESPR's disadvantages are that performance is significantly less than traditional technology, implementing it for large-scale production does require more research. SA-ESPR was found to have higher operating window and production rates as compared to traditional ESPR. The process was found to be more robust and versatile than conventional ESPR. It also opens channels for "green ESPR" and thus has huge potential to be explored for food and food derived products. Similarly, EAPG provides a very good throughput and thus overcomes drawback of conventional ESPR. As these both techniques have a very limited research on food products, a huge scope is there in future on research end which will help in producing encapsulated active compounds on a large scale. These techniques can make it possible to use ESPR at large industrial scale. More intensive research is required on sensory properties of products formed by ESPR. In conclusion, the technique is promising and will possibly attract more and more researchers in future in the field of food and materials science.

Disclosure statement

No conflict of interest is declared by author and coauthors.

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