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Electro-Spinning and Electro-Spraying as Innovative Approaches in Developing of a Suitable Food Vehicle for Polyphenols-Based Functional Ingredients

Mahmoud Ghorbani and Ricardo Santos Aleman

Abstract

With recent advances in medical and nutrition sciences, functional foods and nutraceuticals fortified with natural polyphenols have received a lot of attention from both health professionals and the common population in the last few years since their chemical structure allows them to exert various health effects (e.g., antioxidant, anti-inflammatory, immune, antitumor and prebiotic properties). Nonetheless, there are several hurdles to applications of polyphenols in the food system. The most critical hurdle includes polyphenols' tendency to lose their anti-oxidative properties or bioactive functionalities during food processing, as well as inclusion of poly-phenol compounds may impart an astringent or bitter taste, or introduce a degree of brown coloring causing serious sensorial impacts on food products. On this basis, interest has increased in understanding the development of new and efficient food vehicles as delivery systems for polyphenols-based functional ingredients. In this context, one approach that could augment the growth of polyphenols-based functional foods is electro-hydrodynamic processing, as the most versatile method to produce nanoscale fibers or particulates suitable for application in food technology by encapsulation to form nanoscale delivery systems.

Keywords: Polyphenols, Functional foods, Nutraceuticals, Electro-hydrodynamic processing, Electro-spinning, electro-spun fibers, electro-spraying, electro-sprayed particles and core-shell structured fibers or particles

1. Introduction

During the past few years, there has been increasing awareness about health-promoting effects of dietary polyphenols abundant in functional foods (*natural or processed foods that contain known or unknown biologically-active compounds; which in defined, effective and non-toxic amounts, provide a clinically proven and documented health benefit for prevention, management or treatment of chronic disease*) [1] and nutraceuticals (Natural bioactive or chemical compounds that besides offering

a nutritional value provide health-promoting, and disease curing or prevention properties) [2] as functional ingredients to provide a health benefit beyond basic nutrition. Polyphenols are naturally occurring compounds in plants endowed with antioxidant and anti-inflammatory, immune, antitumor and prebiotic properties [3]; widely present in a wide variety of fruit, vegetables, seeds, herbs and beverages in particular in beer, red wine, fruit juice, coffee, tea, cocoa, chocolate and dry legumes and cereal [2, 4–8] and are therefore an integral part of the human diet [9]. From a chemical standpoint, this large family of secondary plant metabolites constitutes a large heterogeneous class of compounds characterized by hydroxylated phenyl moieties [2, 10–12] with more than 8000 identified compounds so far [2, 13–16]. Numerous food matrixes naturally enriched with dietary polyphenols are the most potent sources of plant-derived bioactive compounds eliciting many beneficial health effects in man. Despite their interesting biologic properties, their presence and abundance in nature, chemical instability of polyphenols during processing, handling and storage [17], the low oral bioavailability [3, 8] and rapid fast-pass metabolism of polyphenols might greatly restrict their biologic effects and applications in the functional foods and nutraceuticals [18]. Further, these extracts or their isolated individual compounds have the potential to interact with other compounds in the environment in particular proteins, resulting in formation of sensory characteristics and organoleptic properties in foods and beverages including High Molecular Weight (HMW) brown color [19], flavor and taste attributes like bitterness or astringency [4, 5, 20, 21].

In conclusion, the main drawback to using polyphenols as functional ingredients to develop functional food products and dietary supplements, nutraceuticals is their poor bioavailability and the variable bio-accessibility in the human body and variety of molecular interactions between polyphenols and other food components; however, in order to preserve the structural integrity, polyphenols need to be shielded by a finishing formulation that is, able to protect and to deliver them to the physiologic targets without losing any bioactivity [22]. Encapsulation system applied to polyphenols through the development of micro and nano-sized particle systems, as a reliable tool to overcome the problems related with the direct use of dietary polyphenols in their free form in food matrixes will ensure protection of these bioactive compounds and additionally, functional properties to the final product [23, 24]. The administration of encapsulated polyphenols instead of their free form can overcome the drawbacks of their instability; relieve unpleasant tastes or flavors in food matrixes, and as well improve their bioavailability in gastrointestinal tract (GIT). Numerous encapsulation processes have been developed to encapsulate polyphenols-based functional ingredients each with their own merits and demerits including Ionic gelation, layer-by-layer deposition, extrusion, coprecipitation, coacervation and phase separation, spray/freeze drying, emulsification/emulsion polymerization, inclusion complexation, liposome entrapment, fluidized bed coating, supercritical fluid, etc. [22, 25]. However, polyphenols are oxidized easily due to light, heat and oxidant; therefore, chemical instability of these compounds is the major constraint to encapsulation through the processes stated above since they mostly require heating and/or pressure, and the use of strong and toxic organic solvents or expensive equipment [26]. In this regard, electro-hydrodynamic processing referring to the dynamics of electrically charged fluids has emerged as an Innovative and environmentally friendly alternative technology for encapsulation that needs neither temperature nor expensive equipment; therefore, heat-sensitive compounds such as dietary polyphenols may be successfully processed and also, the use of organic solvents can be avoided by adjusting some processing conditions (i.e., use of molten polymer).

In this chapter the drawbacks related to the incorporation of dietary polyphenols as possible functional ingredients in food formulations and novel strategy to

improve their efficiency is discussed; starting from bio-accessibility and bioavailability of polyphenols, continuing to the chemical structure of polyphenols, nature of food matrix as well as interaction with other food constituents and also food processing influencing their stability and, consequently their availability and concluding to consider electro-hydrodynamic processing as novel strategy to improve delivery efficiency and controlled release of polyphenols.

2. Bio-accessibility and bioavailability of dietary polyphenols

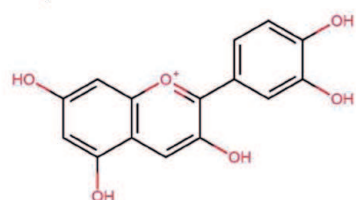
Polyphenols, a class of chemical compounds consisting of one or more hydroxyl groups (OH) attached directly to at least two phenyl rings lacking nitrogen-based functional group in their most basic structural expression are plant secondary natural metabolites, ubiquitous in all vascular plants arising biogenetically from either the shikimate derived phenylpropanoid and/or the polyketide pathway(s) [27–29]. Polyphenols are classified into diverse classes on the basis of their chemical structures and/or the attachment of hydroxyl groups to the aromatic rings structure while the main classes of polyphenols consist of flavonoids, phenolic acid, tannins (phenolic polymers), phenylethanoid [30], stilbenes and lignans [4, 27, 31–34].

Flavonoids are recognized as one of the largest and most abundant type of polyphenols in the diet that constitute approximately two-thirds of intake. The core structural unit of flavonoids encompasses a common carbon skeleton of diphenyl propane in which two benzene rings (A, B) are linked by a linear three-carbon chain, forming a closed pyran ring with the A benzene ring. Flavonoids (**Figure 1**) are then subdivided into several subclasses based on the central pyran ring's oxidation state that the most important of them follow as: flavonols (e.g. Quercetin, kaempferol), flavones (e.g. luteolin, apigenin), anthocyanins (e.g. cyaniding, pelargonidin), flavanones (e.g. naringenin, hesperetin), flavanols also known as flavan-3-ols (e.g. catechin, epicatechin), and isflavones (e.g. daidzein, genistein) [4, 7, 32, 35, 36].

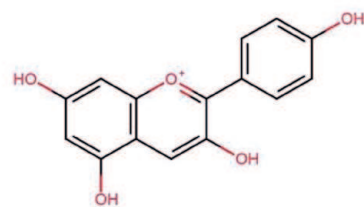
Phenolic acids (**Figure 2**) ubiquitously found in plant materials at varying levels are divided into two sub-classes hydroxybenzoic (e.g., gallic, phydroxybenzoic, vanillic, syringic, and protocatechuic acids) that are often the component of a complex structure like lignins and hydrolyzable tannins, and hydroxycinnamic acids (e.g., p-coumaric, caffeic, ferulic, sinapic and cinnamic acids). Further, decarboxylation of benzoic acid and phenylpropanoid derivatives leads to the formation of simple phenols namely, phenol, o-cresol, 4-ethylphenol, guaiacol, 4-vinylguaiacol and eugenol [4]. Some phenolic acids are found in free form in red fruits and vegetables such as strawberries and blackberries, black radish, onions, and tea [35], but hull, bran, and seed contain phenolic acids that in bound form that are released by acid, alkali, and enzyme hydrolysis [7].

Tannins are compounds of intermediate to high molecular weight (500–20,000 Da) [37] and are more extensively hydroxylated [35]. Depending on their structures, tannins are classified into two major groups including hydrolyzable and non-hydrolyzable tannins, also called condensed tannins or proanthocyanidins (PAs). Hydrolyzable tannins (HTs) consist a center of glucose or a polyhydric alcohol partially or completely esterified with simple phenolic acids such as gallic acid (gallotannins) or hexahydroxydiphenic acid (ellagitannins) while condensed tannins are oligomers and polymers of flavonoids, specifically flavan-3-ols [4, 31, 34, 37, 38]. Ellagitannins (e.g., punicalagin [39]) (**Figure 3**) are esters of hexahydroxydiphenic acid and monosaccharide (most commonly glucose) naturally occurred in some fruits (pomegranate, strawberry, blackberry, and raspberry), nuts (walnuts, almonds) and seeds. While ellagitannins are slowly hydrolyzed in

Anthocyanins

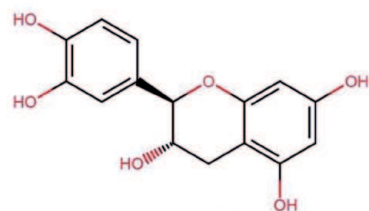


Cyanidins

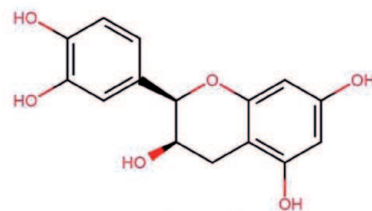


Pelargonidin

Flavanols

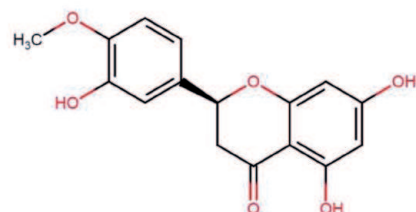


(+)-Catechin

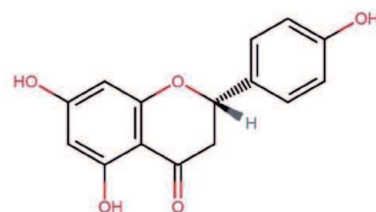


(-)-Epicatechin

Flavanones

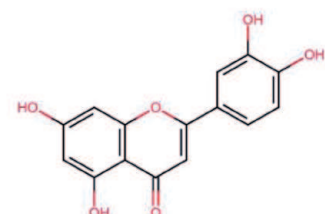


Hesperetin

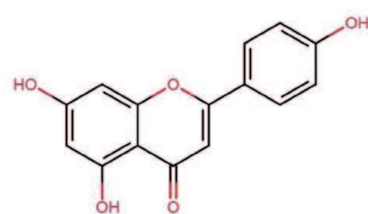


Naringenin

Flavones

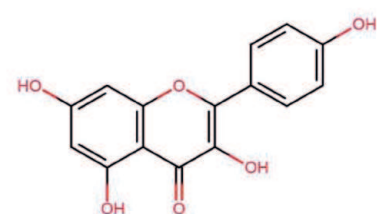


Luteolin

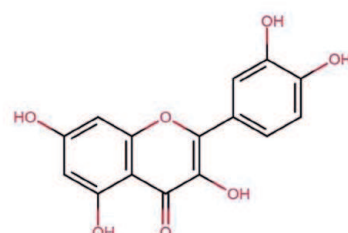


Apigenin

Flavonols

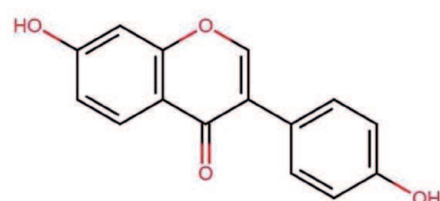


Kaempferol

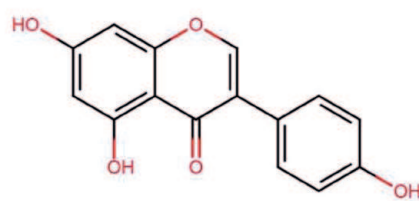


Quercetin

Isoflavones



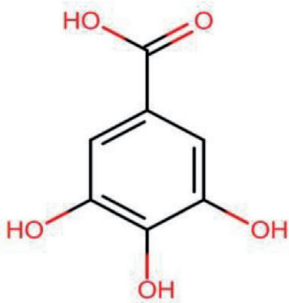
Daidzein



Genistein

Figure 1.
Structures of flavonoids.

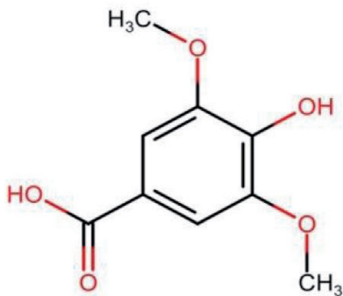
Hydroxybenzoic Acids



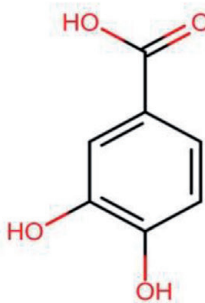
Gallic Acid



Vanillic Acid

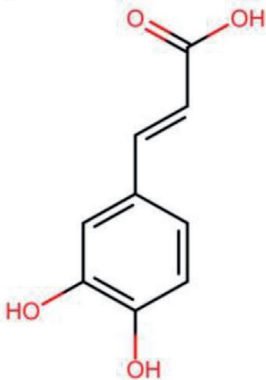


Syringic Acid

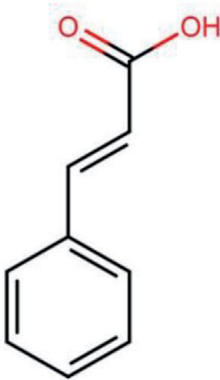


Protocatechuic Acid

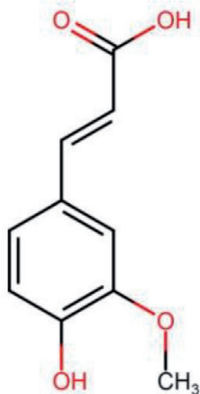
Hydroxycinnamic Acids



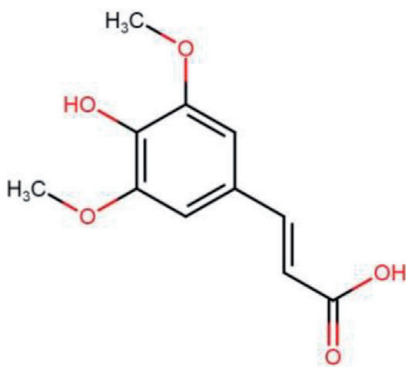
Caffeic Acid



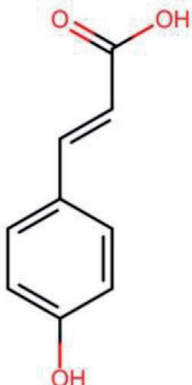
Cinnamic Acid



Ferulic Acid



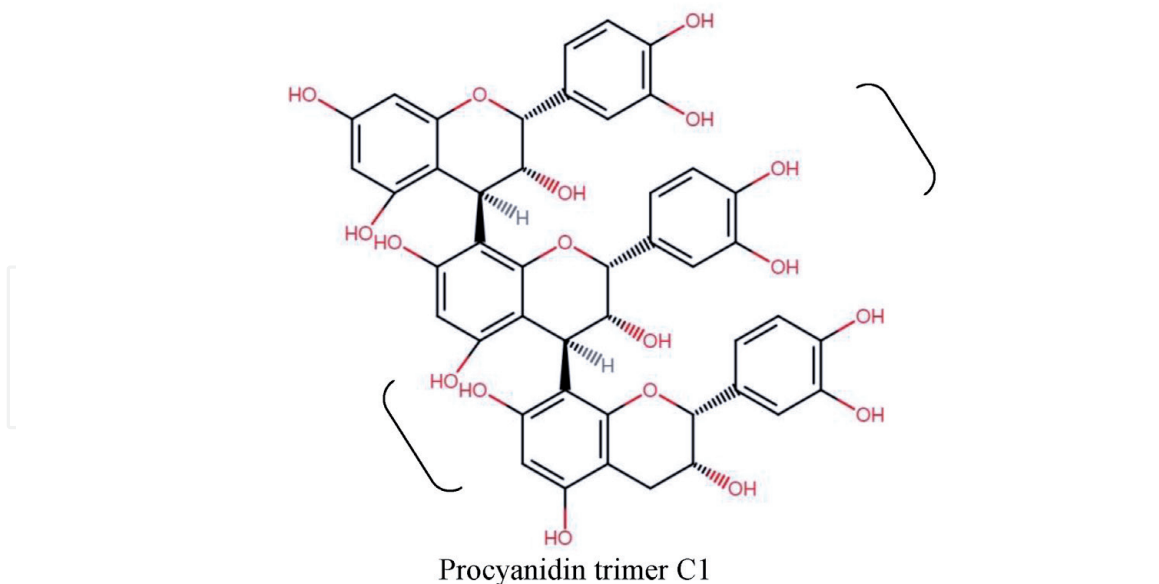
Sinapic Acid



p-Coumaric Acid

Figure 2.
Structures of phenolic acids.

Proanthocyanidins (Condensed Tannins)



Hydrolyzable Tannins- Ellagitannins

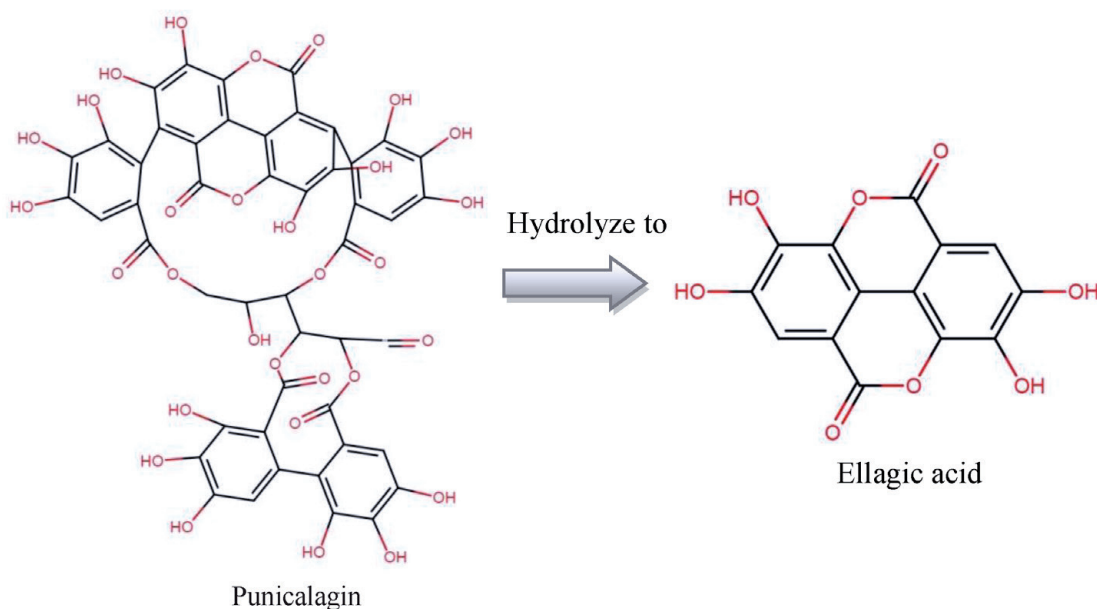
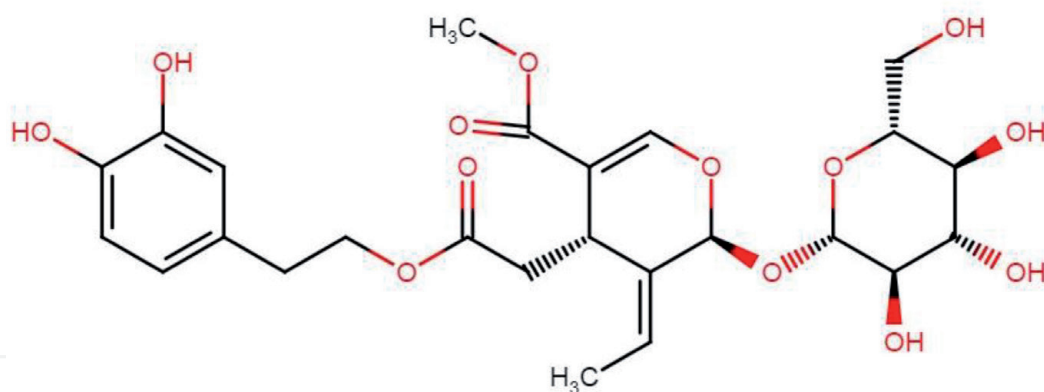


Figure 3.
Structures of tannins.

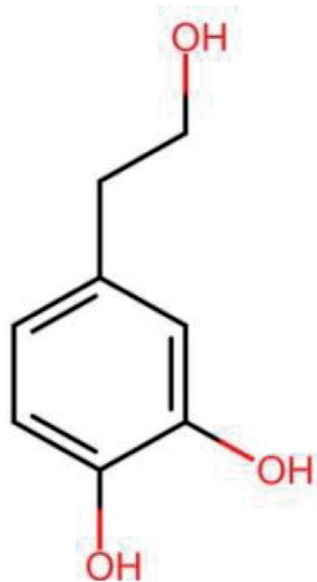
the digestive tract releasing the ellagic acid molecule, the main determinant of the physicochemical properties and biologic activity is their chemical structure [39]. Based on the hydroxylation pattern of A- and B-rings, non-hydrolyzable tannins or PAs can be divided into procyanidins (e.g. procyanidin trimer C1), propelargonidins and prodelphinidins [4, 37].

Tyrosol, hydroxytyrosol and oleuropein (**Figure 4**) are the prominent types in phenylethanoid class, found mainly in olive leaf and oil [30, 36]. Besides, Rueda et al. reported that the minor values of both tyrosol and hydroxytyrosol present in other edible virgin vegetable oils (argan, wheat germ and sesame) [40].

Stilbenes are a family of hydrocarbons that share with similar chemical structure to flavonoids consisting of two phenyl groups linked by a methylene group (or “methylene”) that occur naturally in either a cis or a trans configuration [32, 33]. Resveratrol, pterostilbene, and piceatannol are primary representatives [4, 41] while resveratrol (3,5,40-trihydroxy-trans-stilbene) (**Figure 5**) presented in the both cis and trans



Oleuropein

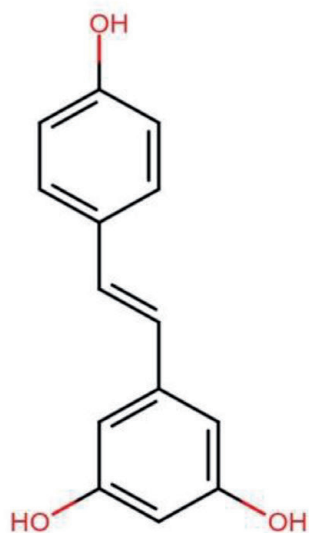


Hydroxytyrosol

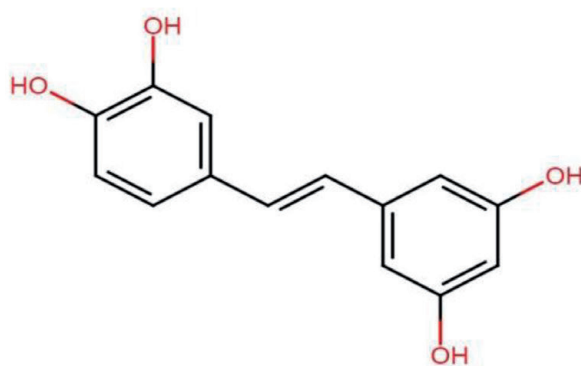
Figure 4.
 Structure of phenylethanoid.

isomeric forms as well as its derivatives including glucosylated, prenylated, methylated, and hydroxylated modifications are the most widely known stilbenes that do are important from a health perspective. Polydatin, also known as piceid (resveratrol-3-O- β -mono-D-glucoside, POLY) is a glucoside of resveratrol in which the glucoside group bonded in position C-3 substitutes a hydroxyl group. This substitution brings about conformational changes of the molecule resulting in changes in the biologic properties. There exist numerous reports suggesting trans-resveratrol to be the more stable form in nature and the most bioactive form of this molecule though upon exposure to UV light, trans-resveratrol (isomeric) can be readily converted to cis-resveratrol (isomeric) and also been unstable when exposed to high pH [42]. Stilbenes are reported to be present in grapes, berries, peanuts and red wine [4, 31, 32, 42].

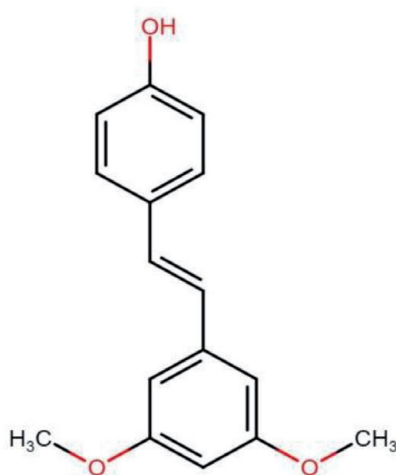
Secoisolariciresinol and matairesinol (**Figure 6**) characterized by the union of two phenylpropanoid (C₆C₃) units with β - β or C₈-C₈ linkages are the major types of lignans [4, 28]; being present in the free form or glycosidically linked to different fiber-associated polyphenols [30]. Flaxseed [4], rye bran and flour, various berry fruits [43], legumes, vegetables, black and green tea [43] are sources of secoisolariciresinol and matairesinol [32].



Resveratrol



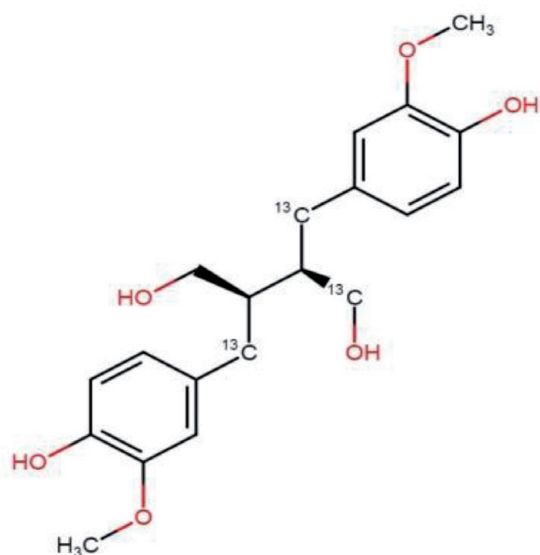
Piceatannol



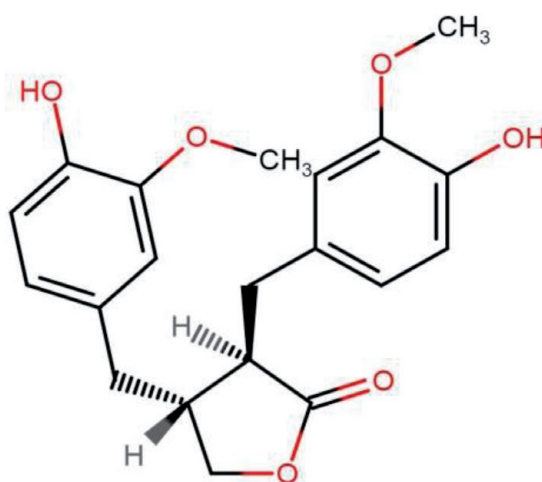
Pterostilbene

Figure 5.
Structures of stilbenes.

These compounds are potent antioxidant compounds able to counteract oxidative stress and chronic inflammation that could be separated or recovered from food waste and agricultural by-products [35, 44]; thus, this group of natural products could be used as additives and functional ingredients in the novel



Secoisolariciresinol



Matairesinol

Figure 6.
Structures of lignans.

functional foods and beverages [45]. There are numerous *in-vitro* and *in-vivo* studies that have proved, poly-phenol intake is associated with a wide spectrum of potential biologic activities related to health-promoting properties including; anti-inflammatory, antioxidant, pro-DNA repair, anti-helminthic, anti-microbial, anti-viral, insecticidal, anti-cancer, anti-aging, and anti-radiation effects [36]; it is known as a preventive for the certain cancers, cardiovascular diseases, type 2 diabetes, osteoporosis, pancreatitis, gastrointestinal problems, lung damage, and neurodegenerative diseases [11, 46–51]. Due to these healthy characteristics, there is increasing interest in the development of new product with enhanced potential health-promoting action while the effectiveness of polyphenols depends on preserving the stability, bioactivity, and bioavailability of the active ingredients.

Once ingested, polyphenols are metabolized by the human body as xenobiotic compounds which may undergo several biologic processes limiting their potential uptake by humans [10, 52, 53]. However, putative bioactivity and bio-efficacy [18, 54, 55] of dietary polyphenols are therefore strictly associated with related to the concepts of bio-accessibility and bioavailability. In this regard, to exploit

the real biologic potential effect it is crucial to know the quantity of polyphenols properly released from food matrix during gastrointestinal digestion, which is the potential absorption of polyphenols to be available for subsequent metabolic pathways. This parameter is known as bio-accessibility, which can be defined as: “the fraction of a bioactive compound within the food matrix where it is contained, that has the potential to be bio-available and reach systemic circulation; this means that it has been released from the food matrix by the action of digestive enzymes [46, 52, 56, 57]. Potentially, bioavailability refers to the fraction of the bio-accessible ingested nutrient, either parent compound or active metabolite that reaches the systemic circulation and becomes available at the site of the action where it can exert biologic effects [46, 52, 58, 59]; in the case of polyphenols, this is the amount of polyphenols which can be absorbed and exert effects on specific tissues. As per this principle, bioavailability is a process that depends on the intrinsic and extrinsic factors of the host; this means that, the process depends on the food matrix ingested and the gastrointestinal conditions within the individual. Investigations have revealed which the bioavailability of polyphenols contains seven main digestive processes: (1) the release of polyphenols from food matrix (bio-accessible polyphenols); (2) changes in polyphenols during gastric and small intestine digestion conditions; (3) the cellular uptake of aglycones and conjugated forms of polyphenols; (4) microbial metabolism conjugated non-bio-accessible fraction polyphenols by the colonic micro-biota; (5) phase I (oxidation, reduction and hydrolysis) and particularly phase II (conjugation) bio-transformations in the enterocytes and then the hepatocytes, followed by generating methyl, glucuronide and sulfate derivatives; (6) transit to systemic circulation and tissue distribution; (7) urinary excretion or excretion back into the gut via bile and pancreatic juices [52, 57, 60]. Numerous studies have pointed out that the bioavailability of polyphenols is rather low and the magnitude of the relative urinary excretion of the intake fluctuates from 0.3% for anthocyanins to 43% for isoflavones that demonstrates the great variability in the bioavailability from one poly-phenol to another and the most abundant dietary poly-phenol was not necessarily the one leading to the highest levels of active metabolites in plasma [34, 51, 61]. Consequently, to explore and to determine the mechanisms of action of dietary polyphenols and their role in disease prevention, it is crucial to understand the factors that constrain bio-accessibility and bioavailability of polyphenols, some related to the food (e.g., chemical structure of the compound, food matrix, food processing and dose) while others depend on the individual (e.g., gastric emptying, intestinal transit time, composition of the micro-biota) [58, 62, 63]. However, the discussion of factors influencing the bio-accessibility and bioavailability of polyphenols will focus on food related factors such as the polyphenols’ chemical structure, the nature of the food matrix and food processing, since these are the first hurdles that polyphenols face prior to absorption.

The interested reader may consult some of the accounts of the concentration and bio-accessibility of poly-phenol compounds with potential antioxidant activity as affected by simulated *in vitro* digestion; for a more detailed description, see the references [64–68].

2.1 Factors leading to degradation of polyphenols and low bioavailability

Dietary polyphenols to exert their health-promoting effect need to endure the food processing conditions; second, could be released from the food matrix and become bio-accessible in the gastrointestinal tract, and then undergo metabolism and reach the target tissue of interest. As a result, chemical structure of polyphenols, nature of food matrix as well as interaction with other food constituents in

particular proteins, lipids and carbohydrates and food processing play a significant role upon coming to bio-accessibility and bioavailability of polyphenols, since they represent the first step in the challenging journey of well-known dietary polyphenols reaching the target tissues.

2.1.1 Chemical composition and structure of polyphenols

Dietary polyphenols has been a most exotic topic in modern food chemistry not only as structural diversity and major plant secondary metabolites, but also as compounds that express a wide range of applications in various aspects of commercial as well as general public interests [27]. The importance of their molecular structure lies in the fact that the molecular size, the parent structure, degree of polymerization or glycosylation, solubility, hydrophobicity, isomer configuration and conjugation with other phenolics [5, 34, 60, 69, 70] have a strong impact on their bio-accessibility and bioavailability. Most of the polyphenols, especially those containing adjacent dihydroxyl groups (e.g. catechins and procyanidins) are especially prone to polymerization and loss through oxidation [71]. Relatively, simple phenolic derivatives such as phenolic acids (e.g., gallic acid, caffeic acid, vanillin, and coumaric acid) and flavonoids including isoflavones are readily absorbed through the gut tract that are followed by catechins, flavanones, and quercetin glucosides [34]. On the contrary, proanthocyanidins which are compounds of high molecular weight are very poorly absorbed as well as galloylated tea catechins and the anthocyanins [34, 60]. Among the various poly-phenol compounds, reported bioavailability is so highly variable that the highest bioavailability has been reported for isoflavones, followed by flavanols, flavanones and flavonol glycosides, while the proanthocyanidins, flavanol gallates and anthocyanidins are the most poorly absorbed [34, 49, 51, 61, 72].

2.1.2 Food matrix

Food products fortified with dietary poly-phenol rich extracts may lead to changes in the nutritional, chemical and rheological properties of the fortified food. Apart from potential biologic activities related to health-promoting properties, when included in a food product depending on the type of extract, the poly-phenol compounds may impart an astringent and/or bitter taste, or introduce a degree of brown coloring [5, 21, 53, 73, 74]. Concerning taste, PAs resulting from oxidative reactions are mostly responsible for some unpleasant organoleptic properties such as astringency and bitterness [51, 70, 71]. "*Astringency is a tactile sensation defined as dryness, tightening and puckering sensations perceived in the oral cavity during the ingestion of astringent molecules, mainly tannins, alums and some metal ions*" [74]. Concerning color, it is worth to note that anthocyanins are one of the most important natural pigments though they represent a problem owing to their high instability [74].

Polyphenols possess the ability to interact, both with food matrix constituents in particular carbohydrates, lipids and proteins, as well as with biologic compounds, namely proteins. All these interactions can affect the accessibility and availability both of polyphenols and other compounds as well as organoleptic properties of fortified food products and consumer acceptance. Polyphenols interact mostly to components of food matrix through non-covalent hydrophobic interactions but in the cases of interactions between polyphenols and proteins or/and carbohydrates, hydrogen bonds also contribute significantly. Nonetheless, some covalent bonds may also occur under certain food processing conditions [74]. Polyphenols form complexes with proteins that can be occurred by non-covalent interaction

(reversible), primarily driven by hydrogen bonds and hydrophobic interactions and covalent interaction (mostly irreversible) after poly-phenol activation either by oxidation, i.e. as quinones, or as carbocations resulting from proanthocyanidin cleavage under hot acidic conditions [38]. As a whole, polyphenols with elevated molecular weight and a more abundance of hydroxyl group, which provide more than one site for interaction reveal a higher affinity to interact with proteins [19, 75]. However, tannins are polyphenols capable of precipitating proteins from aqueous solutions, which are synthesized via the shikimic acid pathway [4]. In terms of non-covalent associations, amino acids (e.g., alanine, valine, isoleucine, leucine, methionine, phenylalanine, tyrosine, tryptophan, cysteine and glycine) may react to tannins through hydrophobic interactions and hydrogen bonds. From a mechanistic point of view, the hydrogen bindings with the carboxyl group of proteins are associated with capability of the hydroxyl groups of polyphenols to donate a hydrogen atom to the nitrogen or oxygen molecule of amino acids (e.g., lysine, arginine, histidine, asparagine, glutamine, serine, threonine, aspartic acid, glutamic acid, tyrosine, cysteine and tryptophan) [19, 38]. In terms of covalent interactions, polyphenols namely tannins can be oxidized under alkaline conditions and reactive oxygen species through enzymatic and non-enzymatic oxidation reactions causing the generation of highly reactive quinone radicals. Following the oxidation step, resultant quinone radical reacts to another quinone radical that is named condensation reaction to form a dimer; a high molecular weight brown color brown color pigment named as tannin that can further interact with amino acids in a polypeptide chain through covalent binding. At the end, these dimers remain highly reactive, in turn they are re-oxidized and cross linked to another polypeptide chain [19, 73]. In this sense, research studies carried-out by Rodríguez-Roque et al., assessed impact of food matrix (water-, milk- and soymilk-fruit juice beverages) on the *in vitro* bio-accessibility of phenolic compounds and hydrophilic antioxidant activity from fruit juice-based beverages and observed that the combination of a blended fruit juice with milk or soymilk could decrease the bio-accessibility of dietary polyphenols due to the formation of complexes among these compounds and proteins of milk and soymilk though the protein precipitation could mask the poly-phenol astringent or bitter taste [67, 68, 76].

Similar to polyphenols strongly associated with proteins, evidence reveals that polyphenols can also form complexes with Carbohydrates (digestible and non-digestible) that are, highly dependent on the molecular weights of the polyphenols, the hydrophilicity of the poly-phenol, and the structure of the carbohydrate (high molecular weight, low solubility, and conformational flexibility) [75, 77]. The associations between carbohydrates and polyphenols can also affect the organoleptic properties [78] but depending on the compounds, these interactions could have positive and negative effects [74]. Besides influencing astringency perception by tannins-proteins interactions, PAs-carbohydrates associations also can lead to an astringency taste and bitterness modulation into fortified foods while tannins have less affinity to carbohydrates than to proteins due to the strong hydrogen bond formation with protein's carboxyl group. Apart from tannins, anthocyanins have the capability to interact with carbohydrates [78], in turn the association with carbohydrates could lead for, on one hand, to a lower extraction yield, and therefore lower color intensity on the final product, and on the other hand to stabilization and enhancement of anthocyanins color [74]. Interestingly, some studies reported that bioavailability of polyphenols could be reduced due to the interaction with polysaccharides [46, 52, 63, 79] while other studies revealed that polysaccharides from human diet could enhance the polyphenols' uptake [78]; however, polyphenols interactions with dietary fibers (non-digestible) are of particularly significant since non-digestible polysaccharides may play role of "poly-phenol carrier" as an

“essential physiologic function” of polysaccharides contributing to the overall health effects of fiber-rich diets [38, 78].

Concerning polyphenols interactions with lipids, only a few studies have investigated the effect of dietary lipid–poly-phenol interactions on taste that have not been of special importance, except in case of plant oils—primarily the one made from olives [46, 74, 78]. In contrary, it should be highlighted that polyphenols can decrease the synthesis of fats and fatty acids in the liver, or delay their absorption in intestines [74, 78]. As reviewed, dietary polyphenols are known to form complexes with macromolecules and to affect on antioxidant values and bio-accessibility that can impair bioavailability of both polyphenols and macromolecules.

Apart from interactions between polyphenols and macromolecules, polyphenols are also known for their strong metal-chelating capabilities. A number of polyphenols (e.g., phenolic acids, flavonoids [79] and also tannins [80]) efficiently chelate trace metal ions, such as Al^{3+} , Fe^{3+} , and Cu^+ [26, 81, 82] that undergo redox cycling reactions and possess the ability to produce reactive radicals such as superoxide anion radical and nitric oxide in biologic systems [83]. This action attributed to the galloyl and catechol groups of poly-phenol compounds results in the diminution intestinal absorption of minerals and trace elements [79, 80].

2.1.3 Food processing

Processing of plant foods and fortified foods with polyphenols-based functional ingredients exert a main impact on bio-accessibility of polyphenols and consequently bioavailability as well as their content and potential antioxidant activity, which depends on the type poly-phenol-rich food, the nature and location of poly-phenol compounds in the food matrix, the intensity and duration of treatment, as well as presence of components that affect absorption efficiency [46, 48, 49, 63, 69, 76, 84–88]. In overall, the bioavailability of polyphenols is determined by their bio-accessibility [89]; this means that the availability as well as accessibility of polyphenols is likely to be affected by the processing methods since food processing brings about changes in chemical composition and structure of polyphenols and in molecular interactions that have an influence on the capacity of a given compound to be extracted during digestion [69] and thus, it may also increase or diminish the bio-accessibility of such health-promoting components [76, 90] that these components may be those that are added to functional foods or are found naturally in foods such as dietary polyphenols. In other words, food processing can give rise to the degradation of polyphenols; thus, reducing their bio-accessible and non-bio-accessible fractions as well can result to chemical or physical modifications in food in such a way that fosters the release and absorption of polyphenols during digestion [69, 70]. This could be explained different effects found according to the intensity and duration of treatment depict two different scenarios including; (1) increase in the bio-accessible and non-bio-accessible contents but no effects on compounds bio-accessibility; (2) decrease in the bio-accessible and non-bio-accessible contents but a modified (increased or decreased) bio-accessibility [69, 70, 76, 90, 91]. In this sense, precise discernment of the concentration of dietary polyphenols reaching the bio-accessible fraction is much more important than the concentration of these compounds in the corresponding food products [70, 76]. Research study carried-out by Rodríguez-Roque et al. [76], assessed impact of processing [high-intensity pulsed electric fields (HIPEF); high-pressure processing (HPP); and thermal treatment (TT)] on the *in vitro* bio-accessibility of phenolic compounds and hydrophilic antioxidant activity from fruit juice-based beverages and observed an improvement up to 38% in the bio-accessibility of individual polyphenols (caffeic and p-coumaric acids from both water-fruit juice beverage (WB) and milk-fruit juice beverage (MB); chlorogenic

and ferulic acids from MB; hesperidin and rutin from all beverages after treatments), mainly by non-thermal methods (HIPEF and HPP). On the contrary, all treatments did not change the bio-accessibility of caffeic and chlorogenic acids from soymilk-fruit juice beverage (SB), as well as naringenin from both WB and MB but diminished the bio-accessibility of ferulic acid from WB. Besides, bio-accessibility of chlorogenic and p-hydroxybenzoic acids from WB were also significantly reduced by HIPEF (between 10 and 11%) and TT (between 11 and 24%) [76]. In another research study by Ribas-Agustí et al., results clearly showed the overall decrease in bio-accessible polyphenols after pulsed electric fields treatments (1.8 and 7.3 kJ kg⁻¹) can be linked to decreased contents in undigested apple, which was probably consequence of their degradation due to process-induced oxidative reactions [91].

Apart from bio-accessibility, polyphenols may lose their antioxidant activities or bioactivity during processing since they are oxidized easily when exposed to high temperature, oxygen and enzymes [46, 92–96], which should be taken into account when processing poly-phenol-rich food matrixes [54, 97, 98]. Dietary polyphenols are degraded at high temperature; thus, thermal treatments diminish the poly-phenol content in polyphenols-rich fortified food and jeopardize the amount of bio-available polyphenols (referring to bio-accessible fraction) due to the loss of thermo-labile phenols or their polymerization [99–101]; however, it has been shown that based on thermal processing technique used, high temperature also gives rise to other modifications turning into positive for the dietary polyphenols bioavailability such as degradation or modification of cell wall polysaccharides, proteins and other matrix factors that may lead to compounds more accessible to absorption [69, 70, 92, 95, 102, 103]. In these cases, the effect of processing can be accounted for by multiplying raw food poly-phenol content by a retention factor (RF), which describes the change in poly-phenol content for a given food due to a given process and was then calculated from the poly-phenol contents of corresponding raw and processed foods and the yield factor value [97, 104]. RFs are calculated according to Eqs. (1) and (2) as follows:

$$\text{Retention factor (RF)} = \frac{\text{concentration of polyphenol in processed food}}{\text{Concentration of polyphenol in raw food}} \times \text{Yield Factor} \quad (1)$$

$$\text{Yield Factor} = \frac{\text{weight of food after processing}}{\text{weight of food before processing}} \quad (2)$$

Thus, the calculated values of RF < 1 indicate a reduced poly-phenol content in the processed food whereas RF = 1 and RF > 1 indicate full retention or an increase, respectively [97]. Most studies evaluating the impact of thermal and non-thermal processing on poly-phenol compounds in terms of quantity and availability have focused on traditional processing technologies such as heat-related thermal treatment (TT) and on novel emerging non-thermal techniques such as HPP, HIPEF and have used to preserve manufactured food or cooking, as well as during the food preparation, i.e. pretreatments on raw material to obtain food. Some interesting studies evaluating the effect of thermal and non-thermal treatments on retention of polyphenols and their antioxidant capacity as well as bio-availability have been conducted [84, 87, 92, 94, 98, 105–108]. The main finding concerning the impact of thermal and non-thermal processing on retention of individual polyphenols is summarized below and in **Table 1**.

Poly-phenol components	Food before processing	Food after processing	Retention Factors					Ref
			Mean RF Value	Min	Max	SD	N	
Hesperetin	Orange juice	Orange pure juice, pasteurized 70°C,30 s	1.03	1.00	1.07	0.03	2	[110, 111]
	Orange juice	Orange pure juice, high-pressure processed (400 MPa/40°C/1 min)	1.27	1.16	1.39	0.11	2	[110, 111]
	Orange juice	Orange pure juice, high intensity pulsed electric fields (35 kV cm-1/750 µs)	1.00	0.96	1.04	0.04	2	[110, 111]
Naringenin	Orange juice	Orange pure juice, pasteurized 70°C,30 s	0.91	0.84	0.99	0.07	2	[110, 111]
	Orange juice	Orange pure juice, high-pressure processed (400 MPa/40°C/1 min)	1.16	1.13	1.20	0.03	2	[110, 111]
	Orange juice	Orange pure juice, high intensity pulsed electric fields (35 kV cm-1750 µs)	0.9	0.87	0.93	0.03	2	[110, 111]
Myricetin	Strawberry juice	Strawberry pure juice, pasteurized 90°C, 30 & 60 s	0.93	0.90	0.95	0.03	2	[112]
	Strawberry juice	Strawberry pure juice, high-intensity pulsed electric fields(35 kV/cm for 1700 µs)	1.00	1.00	1.00	0	1	[112]
Kaempferol	Strawberry juice	Strawberry pure juice, pasteurized 90°C, 30 & 60 s	1.03	1.01	1.05	0.02	2	[112]
	Strawberry juice	Strawberry pure juice, high-intensity pulsed electric fields (35 kV/cm for 1700 µs)	1.05	1.05	1.05	0.00	1	[112]
Ferulic acid	Milk- fruit juice beverage	Milk-fruit juice beverage, high-intensity pulsed electric fields (35 kV cm-1800 µs)	0.89	0.89	0.89	0.00	1	[76]
	Milk- fruit juice beverage	Milk-fruit juice beverage, high-pressure processed (400 MPa/40°C/5 min)	0.89	0.89	0.89	0.00	1	[76]
	Milk- fruit juice beverage	Milk-fruit juice beverage, pasteurized 90°C,60 s	0.81	0.81	0.81	0.00	1	[76]
	Soymilk- fruit juice beverage	Soymilk-fruit juice beverage, high-intensity pulsed electric fields fields (35 kV cm-1800 µs)	0.82	0.82	0.82	0.00	1	[76]
	Soymilk- fruit juice beverage	Soymilk-fruit juice beverage, high-pressure processed (400 MPa/40°C/5 min)	0.81	0.81	0.81	0.00	1	[76]
	Soymilk- fruit juice beverage	Soymilk-fruit juice beverage, pasteurized 90°C,60 s	0.66	0.66	0.66	0.00	1	[76]

Poly-phenol components	Food before processing	Food after processing	Retention Factors					Ref
			Mean RF Value	Min	Max	SD	N	
p-coumaric acid	Water-fruit juice beverage	Water-fruit juice beverage, high-intensity pulsed electric fields fields (35 kV cm-1800 μ s)	0.87	0.87	0.87	0.00	1	[76]
	Water-fruit juice beverage	Water-fruit juice beverage, high-pressure processed (400 MPa/40°C/5 min)	0.91	0.91	0.91	0.00	1	[76]
	Water-fruit juice beverage	Water-fruit juice beverage, pasteurized 90°C,60 s	0.78	0.78	0.78	0.00	1	[76]
p-hydroxybenzoic acid	Water-fruit juice beverage	Water-fruit juice beverage, high-intensity pulsed electric fields fields (35 kV cm-1800 μ s)	0.69	0.69	0.69	0.00	1	[76]
	Water-fruit juice beverage	Water-fruit juice beverage, high-pressure processed (400 MPa/40°C/5 min)	0.82	0.82	0.82	0.00	1	[76]
	Water-fruit juice beverage	Water-fruit juice beverage, pasteurized 90°C,60 s	0.64	0.64	0.64	0.00	1	[76]
Chlorogenic acid	Fruit juice-soymilk beverage	Fruit juice-soymilk beverage, high-intensity pulsed electric fields fields (35 kV cm-800 & 1400 μ s)	0.91	0.85	0.98	0.06	2	[113]
	Fruit juice-soymilk beverage	Fruit juice-soymilk beverage,, pasteurized 90°C,60 s	0.81	0.81	0.81	0.00	1	[113]
Sinapic acid	Fruit juice-soymilk beverage	Fruit juice-soymilk beverage, high-intensity pulsed electric fields fields (35 kV cm-800 & 1400 μ s)	0.99	0.99	1.00	0.00	2	[113]
	Fruit juice-soymilk beverage	Fruit juice-soymilk beverage,, pasteurized 90°C,60 s	0.87	0.87	0.87	0.00	1	[113]
Coumaric acid	Fruit juice-soymilk beverage	Fruit juice-soymilk beverage,, pasteurized 90°C,60 s	0.97	0.97	0.97	0.00	1	[113]
Apigenin	Fruit juice-soymilk beverage	Fruit juice-soymilk beverage, high-intensity pulsed electric fields fields (35 kV cm-800 & 1400 μ s)	0.89	0.79	1.00	0.1	2	[113]
	Fruit juice-soymilk beverage	Fruit juice-soymilk beverage,, pasteurized 90°C,60 s	0.69	0.69	0.69	0.00	1	[113]

Poly-phenol components	Food before processing	Food after processing	Retention Factors					Ref
			Mean RF Value	Min	Max	SD	N	
Clorogenic acid	Apple, Whole unpeeled	apple, high-intensity pulsed electric fields fields (0.4 kV cm ⁻¹ , 5 pulses (0.01 kJ kg ⁻¹ , 20 µs total treatment time)	0.88	0.75	1.02	0.13	2	[92, 114]
	Apple, Whole unpeeled	Apple, high-intensity pulsed electric fields fields (2.0 kV cm ⁻¹ , 35 pulses (1.8 kJ kg ⁻¹ , 140 µs total treatment time)	0.58	0.58	0.58	0.00	1	[114]
	Apple, Whole unpeeled	Apple, high-intensity pulsed electric fields fields (3.0 kV cm ⁻¹ , 65 pulses (7.3 kJ kg ⁻¹ , 260 µs total treatment time)	0.33	0.33	0.33	0.00	1	[114]

Table 1.
Showing retention factors for individual polyphenols in foods and beverages with the processing technologies as following: Pasteurization, High-intensity pulsed electric fields and high-pressure processed- Analysis by chromatography after hydrolysis [56] & Yield factor values collected based on USDA National Nutrient Database for Standard Reference, Release 23 [109].

3. Encapsulation of polyphenols with electro-hydrodynamic techniques

The incorporation of polyphenols as possible functional ingredients in fortified food products, in particular due to their antioxidant capacity is becoming a growing area of research for the food industry. Nevertheless, their application as bioactive components is often hindered by their poor stability and solubility during food processing and storage, burst release and their low bioavailability or chemical instability when exposed to the conditions of the upper GIT which can significantly compromise their envisioned biologic benefits [115–117]; however, in order to preserve the structural integrity, polyphenols need to be sheltered by a finishing formulation that is, able to protect and to deliver them to the physiologic targets without losing any bioactivity [22, 26, 118].

An attractive approach to avoid the degradation of polyphenols is the process of encapsulation, which is referring to a process that involves the entrapment of an active ingredient through a polymeric matrix that seeks to increase stability and to improve bioavailability and the controlled release of bioactive agents through protecting these compounds from adverse environmental conditions or from the GIT (e.g., stomach acid) [117, 119]. Even though recent trends in the field of encapsulation have been focused on the development of techniques to encapsulate food bioactive ingredients such as polyphenols each with their own merits and demerits including Ionic gelation, layer-by-layer deposition, extrusion, coprecipitation, coacervation and phase separation, spray/freeze drying, emulsification/emulsion polymerization, inclusion complexation, liposome entrapment, fluidized bed coating, supercritical fluid, etc. [22, 25, 117], the application of these techniques is restricted owing to the mostly require heating and/or pressure, and the use of strong organic/non-polar solvents or expensive equipment, which result in degradation of heat-sensitive ingredients as well as associated toxicity concerns [120, 121]. In this regard, electro-hydrodynamic (EHD) processing, which refers to the dynamics of electrically charged fluids [120, 122] has emerged as an attractive alternative technology for encapsulation that needs neither temperature nor expensive equipment; therefore, heat-sensitive compounds may be successfully processed and also, the use of organic solvents can be avoided by adjusting some processing conditions (i.e., use of molten polymer) [120, 123, 124]. Principally, electro-spinning also called “electrostatic spinning” [120, 125] and electro-spraying also known as electro-hydrodynamic atomization (EHDA) [126–128] processes considered as kindred EHD processes [129, 130] are very cost effective, highly flexible and robust techniques, where use a uniform electro-hydrodynamic force to break the liquids into fine jets [121, 124, 128, 130–132]. However, electro-spinning is a drawing process based on electrostatic interactions [133] for papering fibers while electro-spraying is a process of liquid atomization by electrical forces [130] for papering particles. These approaches are promising techniques to fabricate delivery vehicles presenting structural and functional benefits for encapsulation of bioactive ingredients while their use in the field food processing and preservation is considerably less explored. Hence, in following section, focus will be on the current work aim to recognize the prospective of both electro-spinning and electro-spraying techniques for one-step encapsulation of dietary polyphenols, respectively into polymeric fibers and particles of micro-and nano-meter diameters.

3.1 Fundamental of electro-hydrodynamic process encapsulation

The incorporation of dietary polyphenols within polymeric particles and fibers of micro-and nanometer diameters is a promising technique to enhance the performance polyphenols-based functional ingredients in food industry [115, 117]. Micro

and nano-sized particles and fibers could enhance stability, encapsulation efficiency ($\geq 80\%$) [134] and oral bioavailability of polyphenols, as well develop controlled delivery or release [126]; thus, facilitating the development of innovative functional foods. Therefore, a clear and precise understanding of electro-hydrodynamic processes is essential to optimize the operating conditions for the nano-encapsulation of various polyphenols and thus, broadening the potential industrial application in food science. The basic setup of uniaxial electro-hydrodynamic technique generally consists of a high voltage power supply, a spinneret with a metallic needle or capillary tube of small diameter (up to 1 mm) [135], a pumping system, and an electrically conductive collector screen connected to an electrical earth, either can be a flat plate or rotating drum [115–117, 120, 123, 130, 131, 134, 136, 137]. In a typical EHD process, the bioactive agent dispersed in a carrier polymer solution or polymer melt is delivered at a fixed solution flow rate [124, 127, 134] to a capillary spinneret connected to the voltage supply [120, 126] by a pump, which forms a droplet at the spinneret apex. Once the droplet is charged under an applied electrical field to the spinneret, the hemispherical surface of droplet is deformed into a conical shape known as the Taylor cone [132, 138, 139] through the action of two major electrostatic forces including internal electrostatic repulsion of similar charges and the coulombic force of external electric field, which is applied between the spinneret apex and the collector [122–125]. With the increase of electric field strength, more electrical charges accumulate on the surface of suspended droplet, especially until a critical point is reached, where internal electrostatic repulsion eventually overcomes the intrinsic molecular tension forces present at the surface of the droplet at the tip of the Taylor cone; an electrically charged jet of the polymer is then ejected from the tip of the Taylor cone and is driven towards the conductive collector (counter electrode [140]) that is, usually held at earth potential to encourage fibers and particles capture. As the jet takes flight between the spinneret and the collector, it experiences a range of competing instabilities including the surface tension driven Rayleigh-Plateau instability [141, 142] and the electrically driven axisymmetric conducting instability and whipping/bending instability [136, 142] (more correctly described as an expanding helix) [122, 123, 129]. Electro-spun fibers are formed if the degree of molecular chains entanglement in the polymer solution and the solution concentration (directly proportional to viscosity and surface-tention) are high enough, the polymer jet from Taylor cone is stabilized, and elongation occurs in flight in initial linear trajectory and continues at an increased rate after the onset of the so-called “whipping instability” (actually a consistent expanding helix) [123], thereby inhibiting the formation of a filament of discrete droplets while electro-sprayed particles are formed providing the solution concentration is low, the polymer jet is destabilized due to varicose instability and hence, fine particulates are formed. These highly charged aerosols self-disperse in space, thereby preventing droplet aggregation and coagulation as well deposited on the collector as micro- or nano-particles [122, 130, 143, 144]. One important advantage of electro-spinning and electro-spraying is that due to whipping/bending instability of the jet and high surface to volume ratio [143], the evaporation of the solvent occurs at an increased rate during jet flight [123] or by blowing hot air on the extruded filament [120] and no heating is needed, which makes these technologies suitable for dealing with thermally sensitive materials (e.g., polyphenols, probiotic bacteria) [145] as well as the use of organic solvents can be avoided by adjusting some processing conditions [124].

In EHD process, molecular weight of the polymer reflecting the entanglement of polymer chains in solutions and solution concentration (directly proportional to viscosity and surface-tention) have the most effect on the formation bead or fiber morphology from the electro-spinning or spraying process [122, 123, 146]. Depending on the viscoelasticity of the polymer solution, the dominating

instability leads to either electro-spray or electro-spinning [123, 127]. Both techniques work on the same physical principles of the ejection of a continuous jet; however, if the degree of molecular cohesion is below a critical level, particulates are formed from the ejecta and not a continuous fiber. This phenomenon is known Rayleigh-Plateau instability as characteristic of the electro-spraying process which is more commonly achieved with low-viscosity, low-molecular weight or low-concentration polymer solutions [125]. In this context, if Rayleigh-Plateau instability [141] dominates the process and manifests varicose waves on the surface of an EHD jet, the jet breaks up to form highly charged fine particles/beads, dispersed in a radial fashion due to coulomb repulsion. In other word, electro-spray transits to electro-spinning when the viscoelasticity of the polymer solution partially or completely suppresses Rayleigh-Plateau instability resulting in necklace-like beaded fibers or long continuous fibers [129]. The most effective parameters, which affect the fabrication of electro-spun fibers or electro-sprayed particles are divided into parameters related to polymer solvent properties (e.g, conductivity, viscosity, and surface tension), parameters related to the process (e.g., the applied electrical field, solution flow rate, and the distance between the tip of the needle and the collector) and ambient parameters (e.g., temperature, humidity and air flow) [124, 147]. Therefore, by manipulating these parameters, multiple morphologies can be attained and continuous polymeric fibers and beads with diameters ranging from a few nanometers to a few microns can be obtained.

The interested reader may consult some of the accounts of the effect of processing parameters on the properties of electro-spun or electro-sprayed materials; for a more detailed description, see the references [122, 123, 148].

3.2 Methods of electro-spinning/spraying encapsulation

Various strategies are available for encapsulation purposes using electro-spinning and electro-spraying. Direct incorporation of the bioactive food compounds such as dietary polyphenols into the polymeric/bio-polymeric carrier is the most common approach to encapsulate these compounds. Using this path, the bioactive component is randomly distributed throughout the fibers or the particles [144]. In this sense, a number of challenges are available that need to be overcome when developing this type of structures. First of all, many natural biopolymers are poly-electrolytes; having strong intermolecular interactions which need to be overcome for the subsequent formation of electro-spun/electro-sprayed structures [149, 150], as well as a certain fraction of the dispersed component is distributed nearby or on the surface of both electro-spun fibers or electro-sprayed particles which these unprotected species are susceptible to degradation owing to exposure of undesirable external environmental factors [134]. Aceituno-Medina et al. [151] encapsulated quercetin within hybrid amaranth protein isolate (API):pullulan (Pul) ultrathin fibers by using the electro-spinning technique. Their finding revealed that the thermal stability of quercetin decreased upon encapsulation, probably due to the dispersion of this antioxidant. However, a sustained-release of quercetin with a rate of ~ 52% from the API:Pul electro-spun fibers was reported during *in vitro* digestion, which probably corresponded to the amount of bioactive molecules distributed nearby to the fiber surface. Similarly, Blanco-Padilla et al. [152] encapsulated two different concentrations of curcumin (0.05 % & 0.075%) within API and Pul ultra-fine fibers using the electro-spinning technique. Their finding revealed the release behavior of curcumin from the electro-spun fibers during an *in-vitro* digestion process (under simulated gastrointestinal conditions) (pH = 2). The burst release of curcumin from electro-spun fibers was reported 14.5-28.6% during the first 10 min, followed by a more gradual increase up to 28.6-55.8% released at 120 min. Third, the

blend formulations often give rise to burst release of some encapsulated compounds [117, 134]. Fuenmayor et al. [118] investigated two types of highly antioxidant phenolic compounds of very different hydrophobicity included gallic acid (GA) (phenolic acid, water-solubility: $\sim 1.4 \times 10^4$ mg/kg at 23°C) and naringenin (NAR) (flavanone, poorly water solubility: $\sim 1.6 \times 10^1$ mg/kg at 23°C) that were homogeneously incorporated by conventional electro-spinning in ultrafine fibers made of zein (Z) a hydrophobic protein extracted from corn maize. It was reported that release of the loaded polyphenols into aqueous environments is pH-dependent. In the sense, release studies revealed a burst release trend with accumulative release threshold minimum for pH 2 and maximum for pH 7, probably due to pH-dependent differences in the molecular cargo-carrier interactions. Forth, conventional EHD also faces enormous challenges for the encapsulation of hydrophilic bioactive molecules into hydrophobic polymers or the hydrophobic bioactive molecules into hydrophilic polymers [117, 127, 134]. Besides, water is not the ideal solvent for electro-hydrodynamic processing since in comparison with organic solvents, it has a high evaporation temperature and high surface tension [127, 150], as well as the presence of organic solvents can result in the inactivation or denaturation of some hydrophilic bioactive substances [117, 127, 134]. However, other novel approaches such as emulsion or coaxial electro-hydrodynamic process have attracted a great deal of attention due to the fabrication core-sheath structures for encapsulation purpose.

3.2.1 Coaxial electro-hydrodynamic process

Coaxial electro-hydrodynamic is a controlled and one-step technique for encapsulation of fragile compounds such as extracts-rich poly-phenol into core-shell structured nano-fibers/particles using a couple of capillary tube where a smaller one is inserted concentrically inside the larger capillary [124, 144]. Coaxial electro-spinning/electro-spraying overcomes technical limitations of direct incorporation of the polyphenols into the polymer solution by its core-shell design [120, 123, 153] resulted in suppressing the initial burst release [115, 154] and thereby, delivering compounds in a controlled manner [117]. In particular, in the coaxial technology, the active component (core) is fed through the inner capillary spinneret while the polymer solution is extruded through the outer capillary spinneret simultaneously in order to acquire core-sheath structures; thus, the component immiscibility problem is alleviated [149]. One important advantage of coaxial configuration is that coaxial structures can be used to generate multiple core-shell structures [155] which involve one or more additional layers for the bioactive ingredients and the potential to adjust the release kinetics of active component by adjusting the number of layers of the protective shell [121, 127, 150, 156]. In the coaxial configuration, the core liquid containing the food bioactive compounds is pumped through inner capillary spinneret and simultaneously, the shell liquid containing polymeric material is extruded through outer capillary spinneret allowing the formation of a charged compound jet consisting of concentrically co-flowing liquids. And then, core-shell structured particles are formed during the charged compound or coaxial jet with appropriate parameters that is generally known as coaxial electro-spraying [121]; however, core-shell particles transit to fibers with an encapsulated core if the outermost shell polymer solution has sufficient viscoelasticity [129]. This technique is known as co-electro-spinning or coaxial electro-spinning. Compared to uniaxial electro-spun fibers and electro-sprayed particles, the coaxial electro-spun fibers and electro-sprayed particles obtain higher encapsulation efficiency [130], enhanced bioactive protection [141], controlled and tunable release of functional compounds and encapsulation of different compounds in the same structure

allowing their release at different stages [153–155]. Torkamani et al. [157] studied encapsulation of poly-phenolic antioxidants obtained from *Momordica charantia* fruit within zein/gelatin shell core fibers via coaxial electro-spinning. Bitter gourd (*Momordica charantia* L) (BG) fruit is rich in flavonoids and polyphenols making it of certain potential value for use in food and nutraceutical industries. This study dealt with encapsulation of bitter gourd extract within bi-layer zein/gelatin fiber nano-structure as alternative polymer geometry, different than spherical configurations achieved by conventional methods. Their finding revealed that produced coaxial fibers showed higher thermal properties than their zein and gelatin uniaxial fiber counterparts; high encapsulation efficiency and sufficient shelf stability demonstrated the suitability of the coaxial electro-spinning process and the robustness of fabricated fibers which could replace conventional methods such as spray drying or freeze drying, as well as coacervation encapsulation method; coaxial electro-spun encapsulated fibers possessed the potential to be used as stand-alone nutraceutical supplement products or as an ingredient (e.g., filling or edible wrapper) in various food products [157]. Similarly, Yang Mao, et al. [158] investigated ferrulic acid/zein composite fibers prepared using a modified coaxial electro-spinning process to improve drug release profiles. Clearly, results of *in vitro* dissolution tests demonstrated that the fibers from the modified coaxial electro-spinning process exhibited a better drug release performance than those from the single-fluid electro-spinning process in terms of initial burst effect, release period, and tailing-off period compared with those from the blend process [158]. In another study, Yuan Shuai et al. [153] encapsulated curcumin in poly (lactic-co-glycolic acid) (PLGA) micro-particles by an improved coaxial electro-spray process and obtained Core-shell structured micro-particles with designated morphologic characteristics and high drug encapsulation efficiency are obtained in the stable cone-jet mode. Their results demonstrate that coaxial electro-spraying process yields micro-particles with improved drug release profiles in comparison with traditional microencapsulation methods.

3.2.2 Emulsion electro-hydrodynamic process

The emulsion electro-hydrodynamic techniques have been also explored to fabricate core-shell structured fibers or particles using water in oil (W/O) or oil in water (O/W) emulsions which can be developed to encapsulate hydrophilic and hydrophobic compounds, e.g., vitamins, carotenoids, polyphenols, enzymes, peptides, oils, flavors, and probiotics respectively. In this approach, an immiscible liquid containing food bioactive compounds (core material) is firstly stabilized by an emulsifier consist of the original emulsions until a stable emulsion is formed and then electro-hydrodynamic solution is prepared by adding shell polymer into emulsion [128]. The core-shell structured electro-spun fibers or electro-sprayed particles obtain by adjusting the operating parameters (voltage, flow rate, receiving distance, etc). Also, the properties of emulsion (viscosity, droplet size, emulsion stability, etc.) play important roles to ensure the success of emulsion electro-hydrodynamic process [121, 159]. Different from coaxial electro-hydrodynamic that utilize a couple of capillary tube where an inner one is inserted concentrically inside the outer capillary to fabricate core-shell structures, emulsion electro-hydrodynamic processing is utilized to fabricate core-shell structures using a single feeding capillary [121, 144] that the formation of electro-spun fibers and electro-sprayed particles is due to the solidification of polymer and coating on emulsion minimizing the amount of organic solvents used in food systems [121, 159]. Referring to recent studies reveal that the application of such a system can prevent the primary release of ingredients and can achieve targeted delivery and controlled release since the

encapsulated bioactive components need to pass through the core-shell structure matrix prior to entering the surrounding medium during the release process and enhance the encapsulation efficiency, solubility, stability, bioavailability and bioactivity of bioactive compounds [118, 159]. Paximada et al. [160] used emulsion-electro-spraying technique to prepare epigallocatechin-3-gallate (EGCG) as well as a modified lipophilic version of EGCG loaded micro and sub-micron structured bacterial cellulose–whey protein isolate (BC-WPI) particles. Two different catechin, hydrophilic (H-EGCG) or lipophilized (L-EGCG), were encapsulated either on the aqueous or the oily phase of the emulsions and then emulsion was electro-sprayed. Particle size and encapsulation efficiency were highly dependent on the type of EGCG (hydrophilic versus lipophilic) and emulsification method and whether the bioactive compound was added to the oily or aqueous phase. The highest encapsulation efficiency was obtained with lipophilic EGCG, which had been added to the oily phase of the emulsion and emulsified by ultrasound (USLO capsules). The stability of EGCG in USLO capsules was tested under different storage conditions. Overall, capsules prepared with WPI and bacterial cellulose protected EGCG from moisture, heat, and dissolution conditions leading to their potential use to enhance EGCG shelf life when incorporated into foods. However, testing of the capsules in food systems remains to be investigated annual report [160].

4. Conclusions

As reviewed, chemical integrity, retention during processing and matrix interactions are some food-related factors hindering polyphenols bio-accessibility and consequently bioavailability that is, a prerequisite for their bioactivity in humans; however, it is possible to overcome it by entrapping these health-promoting components within polymeric particles and fibers of micro- and nanometer diameters through encapsulation process that entail an enhanced release of dietary polyphenols and/or higher absorption in the gastrointestinal tract, but choosing the most adequate encapsulation matrix, optimal core-to-carrier ratio, and operational parameters need to be performed in order to yield a high-quality product. In the case of dietary polyphenols, electro-spun/electro-sprayed structures can be used as the delivery system in foods to protect them during the processing and storage and to transfer these health-promoting components to the target site in the body as well enhance their bioactive functionalities and mask unpleasant taste, such as astringency of some polyphenols. The key advantage of electro-spinning/spraying process is the absence of heat that is, important for preserving the structure and achieving high loadings of polyphenols upon processing storage and thus, as a novel delivery approach for bioactive compounds, it opens a new horizon in food technology with the possibility of commercialization in the near future.

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