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Historical Perspective

Improving the efficiency of natural antioxidant compounds via different nanocarriers

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ABSTRACT

Encapsulation technology, as a promising approach, has been employed for the protection and controlled release of different bioactive compounds including natural antioxidants; there are restrictions for applying these valuable ingredients in real food products, pharmaceuticals, and cosmetics such as low solubility, low shelf life, difficultly in their packaging and handling, losses due to environmental stresses and food processes, undesirable flavors and odors, untargeted release and instability in various conditions during digestion in gastrointestinal tract. Nanocarriers can be employed to overcome these challenges. There are five groups of nanocarriers based on the principal mechanism/ingredient used to make them for the encapsulation of natural antioxidants titled biopolymeric nanoparticles, lipid-based and surfactant-based nanocarriers, nanocarriers made with specially designed equipment, nature-inspired nanocarriers, and miscellaneous ones. The main goal of this study is to have an overview of role of different nanocarriers in improving the efficiency of natural antioxidant compounds for different purposes. It has been verified that antioxidant-loaded nanocarriers can be applied in many formulations with a higher and controlled release antioxidant activity, which would meet the current needs of consumers' expectations towards clean label products.

Contents

Abbreviations: ANN, artificial neural networks; CD, cyclodextrin; DE, dextrose equivalent; DPPH, 1,1-diphenyl-2-picrylhydrazyl; EDTA, ethylenediaminetetraacetic acid; EGCG, epigallocatechin-3-gallate; EO, essential oil; GIT, gastrointestinal tract; GPE, guabiroba fruit extract; HAT, hydrogen atom transfer; HCAs, hydroxycinnamic acids; IN, inulin; LCNP, liquid crystalline nanoparticle; MCT, medium-chain triglyceride; MD, maltodextrin; NLCs, nanostructured lipid carriers; NP, nanoparticle; ORAC, oxygen radical absorbance capacity; O/W, oil-in-water; O/W/O, oil-in-water-in-oil; PEA, polyethyleneimine; PEG, polyethylene glycol; PEO, polyethyleneoxide; PLGA, poly(lactic-co-glycolic acid); PVA, polyvinyl alcohol; PVP, polyvinylpyrrolidone; ROS, reactive oxygen species; RSM, response surface methodology; SET, single electron transfer; SLNs, solid lipid nanoparticles; SPC, soy phosphatidylcholine; TCEP, (tris 2-carboxyethyl) phosphine; TPP, tripolyphosphate; UA, usnic acid; W/O, water-in-oil; W/O/W, water-in-oil-in-water; WPC, whey protein concentrate; WPI, whey protein isolate.

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1. Introduction

Antioxidants are special compounds capable of either inhibiting or delaying the oxidation processes which occur under the influence of atmospheric oxygen or reactive oxygen species, ROS [1][1]. In the view point of food science, it is defined as a substance in foods when present at low concentrations compared to oxidizable substrates, which significantly prevents or decreases the adverse effects of reactive species including ROS and reactive nitrogen species or normal physiological functions in human [2]. These bioactive compounds have many functions, such as blocking metal radical production, stimulating gene expression and endogenous antioxidant production, repairing damaged molecules, and preventing a variety of diseases [3]. Recently, antioxidants have attracted considerable attention in relation to radicals and oxidative stress, atherosclerosis, cancer, cataracts, prophylaxis and therapy, longevity, Alzheimer, and Parkinson [4]. Different studies suggest that the diseases related to oxidative stress like cancer, coronary heart disease, hypertension, obesity, diabetes and cataract are best minimized by the use of natural antioxidants in vegetables and fruits [5].

Natural antioxidants can be divided into two classes, as shown in Fig. 1: (1) enzymatic antioxidants (2) non-enzymatic antioxidants [4,6]. Enzymes, low molecular weight molecules and enzyme cofactors are produced endogenously [4]. When endogenous antioxidants cannot ensure a rigorous control and a complete protection of the organism against oxidation reactions, exogenous antioxidants will be needed. These types of antioxidants (see Fig. 1), as nutritional supplements or pharmaceutical products, contain active compounds of antioxidant activity [7,8]. Many non-enzymatic antioxidants are obtained from dietary sources which include different groups of chemical classes. Polyphenols are the largest class, as can be seen in Fig. **1**. [4,9,10].

Antioxidants can also be classified into two main groups based on their mode of action: hydrogen atom transfer (HAT) compounds and single electron transfer (SET) components [11].

There are a lot of restrictions for using natural antioxidants in food systems; the most important challenges are low solubility, low shelf life, difficultly in their packaging and handling, losses due to environmental stresses (light, oxygen, high temperature, and pH), and food processes (pasteurization, mixing, baking, sterilization, storage, home preparation, microwaving, boiling, steaming and drying), undesirable flavor of phenolic compounds, untargeted release and instability in various conditions during digestion [12]. Encapsulation methods can overcome the restrictions, as reported by many researchers and producers. Encapsulation technology can provide the best delivery system in food formulations [13–15]. Another important factor is the bioavailability of antioxidants which is the ratio of an ingested antioxidant uptaken by our cells as it can effectively improve our body healthy [16]. When antioxidants are loaded into carriers surrounded by a wall of food-grade materials, incorporated antioxidants are protected from different stresses [12,17]; in addition, release of antioxidants from carriers is also targeted and controlled [18]. According to Assadpour and Jafari [16], nanocarriers can be classified into five groups based on the main mechanism/ingredient used to make them for the food industry. They include lipid-based, nature-inspired, specialized-equipment, biopolymer-based, and other miscellaneous nanocarriers.

In this paper, we will have a systematic overview of different nanocarriers applied for loading natural antioxidant compounds and the results on antioxidant activity of nanoencapsulated bioactives; in addition, some applications and examples in different products will also be covered.

2. Role of different nanocarriers on the antioxidant activity of food bioactives

2.1. Biopolymeric nanoparticles (NPs)

Proteins (e.g., β-lactoglobulin, zein, gelatin, soy proteins, collagen, and albumin) and carbohydrates (e.g., chitosan, pectin, alginate, and other polysaccharides) are two major classes of biopolymers [19,20]. Several review articles have addressed the formulation, production, and characterization of biopolymeric NPs for encapsulation of general bioactive compounds; in particular, phenolic compounds [20,21] and carotenoids [173]. Biopolymeric nanocarriers can be produced through application of a single biopolymer (i.e., lactoferrin nanoparticles, starch nanoparticles, alginate nano-hydrogels, and α-lactalbumin nanotubes) or by combination of two biopolymers with various surface charges (e.g., protein-polysaccharide nano-complexes/conjugates like pectin-whey protein nanocarriers) [22]. Single wall material can't provide all the features of an ideal encapsulation agent. In this content, various approaches have been developed to improve the encapsulation of bioactives, most of which are concentrated on carbohydrate-protein mixtures. For example, by forming a thick viscoelastic film at droplet-emulsion interface, the structural destruction of core material can be prevented [23]. Schematic of nano-encapsulated phenolics by polymeric-based method is presented in Fig. 2**.** Also, Table 1 provides a summary of antioxidant-loaded biopolymeric nanoparticles.

Zein NPs were exploited to nanoencapsulate the antioxidant essential oils (EOs) [24]. According to the results, water solubility of EOs was improved upon incorporation into zein with no inhibitory effect on their free radical scavenging ability. Chuacharoen and Sabliov [25] succeeded in milk fortification with β-carotene-loaded zein NPs. Improved β-carotene stability and antioxidant activity at simulated gastrointestinal (GI) environments was also achieved by β-carotene entrapment in zein NPs [25]. Li et al. [26] worked on preserving the antioxidant activity of (2)-Epigallocatechin-3-gallate (EGCG) through loading into β-lactoglobulin NPs.

The drying behavior of sprayed droplets can be improved by hydrolyzed starches often incorporated as the secondary wall materials. Hydrolyzed starches such as maltodextrin (MD), corn syrup solids, glucose and lactose are benefited from high solubility and low viscosity at elevated concentrations of solids. By forming a dry crust around the drying droplets and enhancing the oxidative stability of encapsulated lipophilic antioxidants, these materials can decline the oxygen permeation through the matrix wall [27,28]. Powders may agglomerate or cake due to glass transition-induced crystallization of carbohydrates which might disrupt the structural integrity of the wall matrix, resulting in the release of some encapsulated lipophilic antioxidants and destruction of their structure [29]. In such cases, application of MDs with relatively higher molecular weights and lower dextrose equivalent (DE) values could improve the physical stability of wall matrix; as high molecular weight MDs possess far higher glass transition temperatures. Modification by whey protein could be another method to resolve the problem of MD [30]. For instance, whey protein isolate (WPI) was partly substituted by MD in the work of Oliveira et al. [30] and the impact of such modification on the features of spray-dried pequi oil and its carotene degradation was addressed. The particles were amorphous, and their treatment with WPI and WPI/MD resulted in more uniform and spherical morphology, enhanced thermal stability and retained oil antioxidative capacity. The WPI/MD treatment led to enhanced γ-carotene protection; whereas WPI system was more successful in protecting the δ-carotene, β-carotene and lycopene, in comparison to the bulk oil. Various biopolymers such as tapioca starch, modified starch and MD have been employed as the wall in spray-drying of β-carotene which exhibited various levels of success [31,32].

In coacervation method, a phase separation occurs for a single or a mixture of polyelectrolytes from a solution followed by deposition of newly-formed coacervate phase around the antioxidant ingredient.

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[1] - Reactive oxygen species

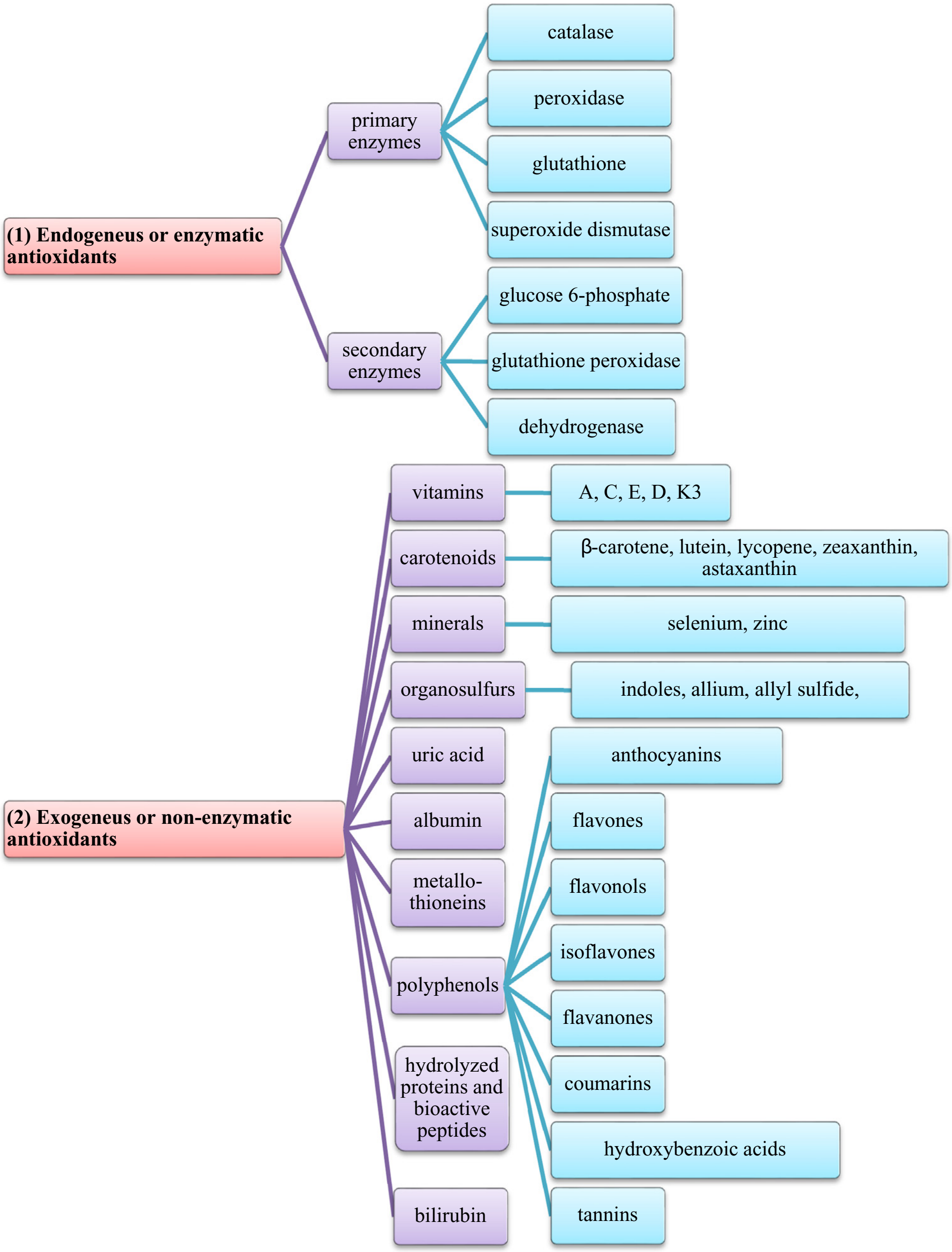

Fig. 1. Classification of natural antioxidants.

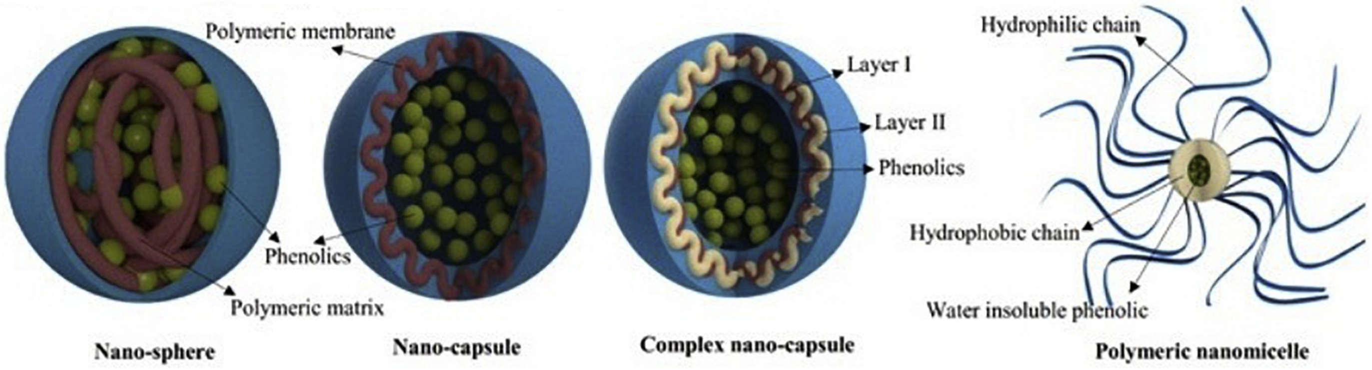

Fig. 2. Schematic representation of nanoencapsulated phenolics by polymeric nanocarriers (typical size <100 nm); reprinted with permission from [116].

Table 1
Selected studies on biopolymeric nanocarriers loaded with antioxidant compounds.

Antioxidants	Nanocarriers	Purpose	Reference
Essential oils	Zein nanoparticles	Increasing the water solubility without hindering their ability to scavenge free radicals	Wu et al. [24]
β-carotene	Zein nanoparticles	Improving stability and enhancing antioxidant activity of β-carotene	Chuacharoen and Sabliov [25]
β-carotene	β-casein micelles	High stability against common food processes	Saiz-Abajo et al. [117]
carotene from pequi oil	WPI and maltodextrin	Enhancing thermal stability and protecting the oil antioxidative capacity	Oliveira et al. [30]
β-carotene	PLA and PLGA	Increasing the stability against destroying during storage	Ribeiro et al. [41]
Capsaicin	Gelatin cross-linked with glutaraldehyde	Improving melting point and antioxidant activity	Wang et al. [35]
Capsaicin	Complex of gelatin and acacia	Increasing antioxidant activity, melting point and improving degradation properties	Jincheng et al. [37]
Curcumin	water-soluble chitosan nanoparticles	Providing high antioxidant and chelating potentials	Yadav et al. [51]
Quercetin	Chitosan nanoparticles	Maintaining the quercetin antioxidant activity	Zhang et al. [52]
Curcumin	Nanofibers based on almond gum/PVA and almond gum/PVA/β-cyclodextrin	Preserving hydrophobic antioxidants	Rezaei and Nasirpour [97]
Curcumin	Chitosan cross-linked with tripolyphosphate	Increasing antioxidant and anticancer activity	Sowasod et al. [42]
Curcumin	β-casein micelles	Increasing the solubility of curcumin and its bioavailability and antioxidant activity	Esmaili et al. [98]
Phenolics, tocopherols, sterols from red fleshed pitaya seed oil	Gum Arabic/maltodextrin Gum Arabic/lactose Sodium caseinate/ Maltodextrin Sodium caseinate/lactose Whey protein/maltodextrin	Increasing antioxidant activity and stability	Lim et al. [46].
(+) catechin and EGCG	Chitosan nanoparticles	Protecting catechin and EGCG from degradation and keeping the antioxidant activity	Dube et al. [49]
EGCG	β-lactoglobulin	Reserving the antioxidant activity	Li et al. [26]
EGCG	Caseinophosphopeptide and chitosan	Stronger free radical scavenging of nanoencapsulated EGCG	Hu et al. [57]
Root extract	PLGA nanoparticles	Higher ability to scavenge free radicals and a higher solubility and stability of bioactives	Kumar and Anand [53]
Quercetin and catechin	PLGA nanoparticles	Enhancing the inhibitory effect on free radicals and the chelating properties	Pool et al. [54]
Quercetin	Chitosan nanoparticles	Improving the bioavailability	Zhang et al. [52]
Phenolic extract of guabiroba fruit	PLGA nanoparticles	Enhancing antioxidant activity	Pereira et al. [56]
Pequi oil and its carotene	WPI, maltodextrin and inulin	Higher thermal stability and improved protective features for the oil antioxidative capacity	Oliveira et al. [30]
Anthocyanins	Alginate and inulin	Increased antioxidants protective properties, improved stability over long storage	Waterhouse et al. [58]
Anthocyanins	WPI/ pectin	Preventing changes in antioxidant activity	Stănciuc et al. [60]
Jujube pulp and seed extracts	Chitosan nanoparticles	Enhancing antioxidant activity and storage stability	Han et al. [118]
Catechin	Chitosan/poly (γ-glutamic acid)	Enhancing the transport and antioxidant activity	Tang et al. [119]

Furthermore, cross-linking of a hydrocolloid shell (by proper chemical or enzymatic cross-linkers like glutaraldehyde or transglutaminase) can improve the coacervate robustness [33]. Depending on the applied polymer diversity, this process can be labeled as simple (single type of polymer) and complex (two or more polymer types) coacervation. Gouin [34] expressed that the coacervation method is a distinguished and highly potential encapsulation approach owing to its high achievable payloads (up to 99%) and controllable antioxidant release by mechanical stress, temperature, or sustained release. Coacervation process was employed by Wang et al. [35] to encapsulate capsaicin inside gelatin through the use of glutaraldehyde cross-linker and vacuum oven drying. Encapsulation led to an improvement in the product melting point and the thermal pyrolysis temperature. Complex coacervation was also applied by Xing et al. [36] to encapsulate capsaicin in gelatin and acacia. Nanoencapsulation involved hydrolysable tannins treatment of encapsulated capsaicin and cross-linking by glutaraldehyde followed by freeze drying procedure. Jincheng et al. [37] used a similar strategy for capsaicin encapsulation; the only difference was vacuum oven drying instead of freeze-dryer. Their fabricated nanocarriers exhibited higher antioxidant activity and melting point (from 75 to 85 °C) as well as lower degradation.

Curcumin was successfully encapsulated in poly(lactic-co-glycolic acid) (PLGA) with stabilizer polyethylene glycol (PEG) by Anand et al. [38] through nano-precipitation followed by freeze drying. The nanocarriers possessed augmented cellular uptake and bioactivity (both *in vitro* and *in vivo*). In a study by Gou et al. [39], curcumin encapsulation was conducted through single-step nano-precipitation and freeze drying techniques. *In vivo* study on the curcumin-loaded nanocarriers showed their substantial antioxidant and anticancer behavior when compared with free curcumin. Solvent displacement was combined with freeze drying method by Tachaprutinun et al. [40] for astaxanthin encapsulation which resulted in proper encapsulation efficiency (98%). Additionally, the freeze-dried astaxanthin-encapsulated nanospheres exhibited acceptable water dispersibility. In the work of Ribeiro et al. [41], solvent displacement as well as freeze drying was employed for production of β-carotene-loaded nano-dispersions through PLA and PLGA biodegradable polymers[2]. The nanoencapsulated β-carotene powder showed higher degradation resistance throughout the storage period; furthermore, it possessed higher antioxidant activity in comparison with non-encapsulated β-carotene. Sowasod et al. [42] reported successful encapsulation of curcumin in chitosan by multiple emulsion/solvent and freeze drying techniques in which tripolyphosphate (TPP) served as cross-linker. The mentioned method yielded an encapsulation efficiency of 72%. The resultant nanocarriers had a spherical shape. Moreover, the antioxidant and cancer cells

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[2] - Biodegradable polymers are the synthetic polymers that are usually biodegradable and made from biological raw materials, but are prepared by chemical methods.

inhibitory behavior of curcumin-loaded nanocarriers were twice the curcumin alone.

According to Hogan et al. [23], whey proteins can effectively serve as wall material to encapsulate the oil soluble food ingredients through spray drying [43–45]. Red fleshed pitaya seed oil can be considered as a promising source of natural antioxidants (i.e. phenolics, tocopherols and sterols). The impact of wall composition on encapsulation and stability of encapsulated red fleshed pitaya seed oil was evaluated by Lim et al. [46]. Six variable matrices were used including gum Arabic/MD DE10, gum Arabic/lactose, sodium caseinate/MD DE10, sodium caseinate/lactose, whey protein/MD DE10 and whey protein/lactose. Sodium caseinate followed by whey protein and gum Arabic were ranked as the most effective wall substances for pitaya seed oil encapsulation. Concerning the stability and antioxidant activity of encapsulated oil, lactose performance was significantly higher than MD.

Nano-gels refer to nanostructured carriers produced through chemical or physical cross-linking of the biopolymer networks [47]. These carriers possess good swelling ability in the suitable solvents. When using water as the solvent, nano-gels are known as “hydrogels” [48]. Several biopolymers including alginate, chitosan, whey proteins and soy proteins can form nano-gel carriers if cross-linked with proper agents. Various synthetic polymers including polyvinylalcohol (PVA), polyethyleneoxide (PEO), polyethyleneimine (PEA), polyvinylpyrrolidone (PVP), and poly-N-isopropylacrylamide can also be employed for nano-gel production; most of which are used for drug delivery purposes. Dube et al. [49] reported catechin and EGCG encapsulation in chitosan–TPP through ionic gelation and sonication combined with freeze drying procedure. Nanoencapsulation efficacy was evaluated by incorporation of reducing agents (i.e. ascorbic acid, dithiothreitol, and (tris 2-carboxyethyl) phosphine (TCEP)) regarding their degradation preserving capability toward catechin and EGCG. Based on the results, nanoencapsulated catechin and EGCG exhibited proper protection in comparison with reducing TCEP and ascorbic acid agents. Furthermore, no variation was seen in the antioxidant activity of catechin and EGCG.

Various antioxidants such as β-carotene, curcumin and anthocyanins have been encapsulated within biopolymer NPs. The resultant soluble nano-complexes exhibited enhanced environmental stability as they were entrapped by strong walls. The antioxidant activity of encapsulated bioactives was weaker than their non-encapsulated peers. This could be assigned to an additional barrier provided by strong complex walls; hence the food ingredients release will be faced with a strong obstacle giving rise to prolonged release and thus lower antioxidant activity [21,50]. Yadav et al. [51] encapsulated curcumin in water-soluble chitosan NPs (<50 nm) and reported high antioxidant and chelating activities (in far less dosage) in the encapsulated samples than free curcumin. Zhang et al. [52] assessed quercetin encapsulation by chitosan NPs to improve its pharmaceutical applications. According to DPPH test results, quercetin-loaded NPs were capable of reducing the stable DPPH radicals to the yellow diphenylpicrylhydrazine; their scavenging ability was dependent on the NPs content. Thus it could be concluded that the created complexes managed to sustain the quercetin antioxidant activity. The reducing strength of quercetin-loaded NPs showed good correlation with concentration enhancement. The results also suggested effective encapsulation of quercetin in the chitosan NPs with more profound reducing power.

Hemidesmus indicus root extract encapsulation in PLGA NPs was also studied and the antioxidant behavior of free and encapsulated extract was evaluated using DPPH, superoxide, and hydroxyl radical scavenging assays. The extract encapsulation resulted in dramatic increase in free radical scavenging ability and hence improved therapeutic efficacy when compared with free extract. Such an enhancement might be the consequence of greater bioactives solubility and stability in the extract [53]. Quercetin or catechin-loaded PLGA NPs were prepared by Pool et al. [54] through solvent displacement technique. They also assessed the antioxidant properties in terms of superoxide anion-scavenging, lipid peroxidation and metal chelation activity. The inhibition effects on free radicals and the chelating features of quercetin and catechin showed an improvement upon encapsulation [54].

Considering the key role of ROS-induced oxidative damage in cancer onset [55], reduction potential of free extract of guabiroba fruit (GPE) and GPE-loaded PLGA were addressed by Pereira et al. [56]. In the case of non-cancer cells, free GPE and GPE-loaded PLGA NPs effectively declined the ROS generation. Therefore, PLGA-encapsulated GPE managed to enhance the ROS generation inhibitory impact of GPE by decreasing the required content of extract. Pereira et al. [56] also encapsulated GPE by PLGA NPs through a modified emulsion-evaporation method; which showed promising preservative impacts on the phenolic compounds and their bioactivity. In a study by Hu et al. [57], mono-dispersed NPs were synthesized using bioactive caseinophosphopeptide and chitosan to nanoencapsulate EGCG. The CAA assay was employed to measure the antioxidant activity of nanoencapsulated EGCG which revealed stronger free radical scavenging features of EGCG-loaded caseinophosphopeptide and chitosan NPs compared to free EGCG. Oliveira et al. [30] partially substituted WPI by MD and inulin (IN) and examined its impact on the spray-dried pequi oil and its carotene degradation rate. The obtained particles were amorphous, and their treatments with WPI and WPI/IN resulted in smoother spherical particles. WPI and WPI/MD exhibited a higher thermal stability and improved protective features for the oil antioxidative capacity indicated by β-carotene bleaching assay. The WPI system exhibited improved β-carotene, δ-carotene and lycopene protection in comparison with the bulk oil; whereas the WPI/MD samples showed enhanced protective features for γ-carotene; investigation of WPI/IN was indicative of its improved protective impact for α-carotene.

Waterhouse et al. [58] used alginate and inulin to produce anthocyanin-rich powders with favorable reconstitution features in water or milk employing the blueberry wastes. In comparison to inulin, application of alginate as the encapsulant/drying aid provided enhanced powder yields, increased antioxidant protective properties, improved stability over long storage periods or high temperatures, and higher retention of total phenol and total anthocyanin content at the simulated GI conditions. The total phenol and anthocyanin contents of powders extracted with no enzymatic treatments were significantly greater than the samples synthesized through enzymatic treatments; which reflects the impact of intrinsic polysaccharide content of blueberry wastes (pectins) on their antioxidant behavior during spray drying process. The biopolymer type (alginate or inulin) can affect the stability of encapsulated polyphenols in a way that alginate can cause enhanced emulsifying effect hence exerting higher antioxidants protection (as suggested by total phenol, total anthocyanin and vitamin C contents). At the mentioned conditions, alginate may also stabilize the vitamin C similar to pectins and polygalacturonic acid [170]. This highlights the importance of uronic acid units of alginate. Presence of extra polysaccharides in the blueberry wastes (including pectins) can enhance the glass transition temperature resulting in more complete encapsulation by forming an improved encapsulating network for antioxidants entrapment. Furthermore, polyphenols can form complexes with the blueberry waste substances (e.g., polysaccharides) through inclusion complexation and/or noncovalent interactions [59]. These complexes have a higher thermal stability as compared to pure polyphenols.

Stănciuc et al. [60] addressed the encapsulation of anthocyanins derived from grape skins in WPI and two different WPI-polysaccharide matrices in which DPPH radical scavenging assay was employed to assess the antioxidant activity of samples. A modified Folin-Ciocalteu method was also applied to evaluate total phenolic content, TPC [61]. According to the results, WPI-encapsulated extracts exhibited a lower DPPH radical scavenging activity. It was anticipated as encapsulation process can influence the bioactivity of core substances, in particular when the structures of core and wall are chemically different. Based on the quenching tests and molecular docking computation results, α-lactalbumin and β-lactoglobulin in WPI are highly involved in the tight binding of anthocyanins. Anthocyanins however were successfully encapsulated by a WPI-carbohydrate polymer mixture; encapsulation with pectin produced finer particles and exhibited a significant higher polyphenols and flavonoids entrapment. Both of the studied carriers demonstrated acceptable antioxidant behavior suggesting their potential applicability as the natural antioxidants.

2.2. Lipid-based and surfactant-based nanocarriers

Various classes of lipid-based nanocarriers have been studied for encapsulation of antioxidants [62]. Nanoemulsions, nanoliposomes, nanostructured lipid carriers (NLCs) and solid lipid nanoparticles (SLNs) are the major studied and developed types in the recent years [63]. Antioxidant-loaded lipid-based and surfactant-based nanocarriers are briefly described in Table 2. The nanoemulsion production approaches can be classified into high energy (i.e. high-pressure homogenization and ultrasonication) and low energy (i.e., use of controllable mixture at low stirring known as spontaneous emulsification) methods [18]. Considering the nature of antioxidants, two groups of emulsion systems can be introduced: oil in water (O/W) and water in oil (W/O) nanoemulsions for nanoencapsulation of water-insoluble (lipophilic) and water-soluble (hydrophilic) antioxidants, respectively [12,64].

Mao et al. [65] showed proper environmental stability of β-carotene loaded in WPI-stabilized O/W nanoemulsions. Mehrnia et al. [169] evluated release properties and rheology of nanoemulsions loaded with crocin, as a natural carotenoid of saffron. In another study, nanoemulsions were prepared by whey protein concentrate (WPC) and Tween 80 and to nanoencapsulate curcumin in medium-chain triglyceride (MCT) oil droplets [66]. The loaded curcumin showed enhanced stability and bioavailability in comparison with non-encapsulated ones [66]. Sessa et al. [67] examined the antioxidant properties of grape marc polyphenols encapsulated in O/W nanoemulsions (sunflower oil and palm oil) and compared them with the non-encapsulated extracts using ORAC assay. The antioxidant behavior of encapsulated grape polyphenols was comparable or even greater than non-encapsulated ones suggesting their similar accessibility for the peroxyl radical scavengers, which seemed to be not affected by their inclusion in a lipid phase. Tiyaboonchai et al. [68] applied microemulsions and freeze drying methods to load curcuminoids into SLNs. Under optimal conditions, lyophilized curcuminoids-loaded NPs exhibited the highest antioxidant activity. Furthermore, any alternation in the ingredient contents (e.g. lipid and emulsifier) dramatically affected the curcuminoid loading capacity and bioactivity. *In vitro* release investigations indicated a sustained curcuminoids release (up to 12 h); the product was also physically and chemically stable during 6-month storage.

Mohammadi et al. [69] succeeded in designing a stable and sustained-release nanoencapsulation system to deliver olive leaf phenolics through W/O/W multiple emulsions stabilized by WPC and pectin. Their results showed superior antioxidant properties of encapsulated samples over the non-encapsulated olive leaf extract. Qian et al. [70] loaded β-carotene in orange oil droplets using O/W nanoemulsions which was stabilized by β-lactoglobulin and Tween 20. Their results indicated that β-carotene encapsulated by protein-coated lipid droplets possessed high chemical degradation stability comparied with those encapsulated by non-ionic surfactant coated droplets. Ru et al. [71] managed to encapsulate EGCG by nanoemulsions stabilized with ι-carrageenan and β-lactoglobulin, to synthesize a biocompatible EGCG carrier. Nanoencapsulated EGCG by nanoemulsions (400 nm) exhibited higher *in vitro* antioxidant and anticancer properties than free EGCG [71]. Coenzyme Q10 is an antioxidant compound which was loaded in lecithin and Tween 20-stabilized nanoemulsions [72]. Encapsulation induced no structural alternation in the coenzyme Q10 after 70 days of storage at 21 °C. Also, the obtained nanoemulsions offered a proper and stable dissolving environment for coenzyme Q10.

Nanoliposomes can be used for both hydrophilic and hydrophobic antioxidants [73]. Schematic of nanoencapsulated phenolics by nanoliposomes is shown in Fig. 3**.** In a work by Chen et al. [74], curcumin was successfully loaded in nanoliposomes through thin film and dynamic high-pressure microfluidization technique. Their product manifested proper water solubility, physicochemical stability and sufficient bioavailability when compared with free curcumin. According to Cadena et al. [75], antioxidant properties of typical flavonoids (i.e. quercetin) can be enhanced upon their incorporation in nanoliposomes. Dutta and Bhattacharjee [76] applied nanoliposomes to encapsulate piperine-rich black pepper extracts. Their comparison with pure piperine-loaded nanoliposomes indicated greater encapsulation efficiency (78.6%) for piperine-rich extract nanoliposomes with enhanced antioxidant properties and improved storage stability.

Ramezanzade et al. [77] extracted bioactive peptides through enzymatic hydrolysis of rainbow trout skin gelatin and encapsulated them in chitosan-coated liposomes. The molecular weight of encapsulated product was < 30 kDa with enhanced stability which could be attributed to peptide release under laboratory conditions. They also reported that chitosan-coated nanoliposomes can sustain the antioxidant features [77]. Mosquera et al. [78] succeeded in encapsulation of the sea bream-extracted collagen peptidic fractions in nanoliposomes composed of partly-purified soy phosphatidylcholine (SPC). They reported that ABTS radical scavenging ability of encapsulated peptide fractions remained constant after 8 days; however, the ABTS radical scavenging ability of free peptide fractions significantly decreased by around 42.2%. At t=0, the antioxidant capacity of encapsulated sample was about two times more than its free counterpart which could be assigned to the ABTS radical scavenging capacity exhibited by SPC, as its antioxidant activity is far less than the free peptide fractions.

Numerous review articles have addressed formulation, preparation and characterization of SLNs [79–81]. The mentioned approach can decline the leakage of entrapped bioactives and enhance their preservation. However, regarding the crystallization consequence of solid lipids, several researches have optimized this technique through substitution of solid phase with a solid-liquid lipid mixture giving rise to the so-called NLCs [82]. Enzymatic antioxidant incorporation in SLNs can protect them from proteolysis as well. Qi et al. [83] loaded CAT in SLNs obtained through high-purity SPC. Their results confirmed the proteolysis protection capability of SLNs reflecting the promising ability of SPC in functional foods delivery and preservation.

NLCs have been successfully used to load many water insoluble phenolics and antioxidants to enhance their bioavailability. As an example, lutein was loaded into NLCs by Mitri et al. [84]. The obtained nanocarriers succeeded in lutein protection against the environmental stresses. Solvent diffusion method was employed to produce NLCs from Tween 20 and a mixture of palmitic acid and corn; which was then used to load β-carotene [85]. The resulted ultra-small NLCs exhibited superior β-carotene retention leading to its enhanced bioavailability through protection by lipid NPs; thus, NLCs could be regarded as a promising β-carotene nanocarrier in the food industry [85].

Niosomes are widely applied in the drug delivery, serving as the alternative to liposomes with additional advantages. Specifically, they are becoming popular in the field of topical drug delivery due to their outstanding characteristics. Abaee and Madadlou [86] fabricated α-tocopherol-carrying niosomes and charged it into a transglutaminase-cross-linked whey protein solution that was subsequently gelled with glucono deltalactone. Encapsulation efficiency of a-tocopherol within niosomes was 80% and encapsulation did not influence the radical scavenging activity of a-tocopherol. The DPPH scavenging activity of free α-tocopherol and α -tocopherol-loaded niosomes were indifferent (30.0% and 28.7%, respectively). In this work, the niosomal whey protein cold-set hydrogels were fabricated as a novel functional food formulation considering high nutritional value of whey proteins and superb delivery potential of niosomes and to preserve niosomes from adverse environmental and/or gastrointestinal conditions. The tocopherol-loaded niosomes may, however, be released within the upper small intestine and facilitate vitamin adsorption through epithelial cell membranes.

Table 2
Some studies on lipid-based and surfactant-based nanocarriers loaded with antioxidant compounds.

Antioxidants	Nanocarriers	Purpose	Reference
β-carotene	O/W nanoemulsions stabilized by whey protein isolate (WPI)	Good stability against environmental stresses	Mao et al. [65]
β-carotene	O/W nanoemulsions of orange oil droplets stabilized by β-lactoglobulin and Tween 20	Increasing the stability of β-carotene to chemical degradation	Qian et al. [70]
β-carotene	NLCs	Protection and increasing the bioavailability of β-carotene	Hejri et al. [85]
β-carotene	Nanoemulsions with sodium caseinate	Improving the stability and *in vitro* bioaccessibility	Yi et al. [120]
Curcumin	Nanoemulsions of MCT oil droplets with whey protein concentrate (WPC) and Tween 80	More stability and bioavailability	Sari et al. [66]
Curcumin	Nanoliposomes	Increasing water solubility, physicochemical stability, and adequate bioavailability	Chen et al. [72]
Curcumin	Nanoliposomes	Enhancing the gastrointestinal absorption, bioavailability and plasma antioxidant activity	Takahashi et al. [121]
Curcuminoids	SLNs (stearic acid, glyceryl monostearate, and Poloxamer 188)	Increasing antioxidant activity, prolonged release and maintaining the physical and chemical stability during storage period	Tiyaboonchai et al. [68]
Grape marc polyphenols	Sunflower oil and palm oil-based O/W nanoemulsions	Higher antioxidant activity in case of ORAC	[122]
Olive leaf phenolic extracts	W/O/W emulsions of soybean oil stabilized by WPC and pectin	Increasing the antioxidant activity, stability and sustained-release	Mohammadi et al. [69]
EGCG	Stabilized nanoemulsions by ι-carrageenan and β-lactoglobulin	Enhancing *in vitro* antioxidant and anticancer activity	Ru et al. [71]
Quercetin	Nanoliposomes	Enhancing antioxidant capabilities	Cadena et al. [75]
Quercetin	Nanoliposomes (cholesterol and egg PSC)	Enhancing bioavailability	Priprem et al. [123]
Quercetin	SLNs (glyceryl monostearate and soy lecithin)	Enhancement of gastrointestinal absorption	Li et al. [175]
Quercetin	NLCs (linseed oil (liquid lipid)), glyceryl monostearate (solid lipid)	Improving antioxidant activity	Huang et al. [124]
Quercetin	NLCs (Imwitor 900 K, medium chain triglycerides(	Improving bioaccessibility and stability	Aditya et al. [125]
Quercetin	Nanoliposomes	Improving storage stability and antioxidant activity	[126](
Bioactive peptides from enzymatic hydrolysis of rainbow trout skin gelatin	Chitosan-coated nanoliposomes	Maintaining the antioxidant activities	Ramezanzade et al. [77]
Sea bream-extracted collagen peptide fractions	Nanoliposomes composed of partly-purified soy-derived phosphatidylcholine	Maintaining the radical scavenging ability	Mosquera et al. [78]
Catalase	SLNs	Protecting against proteolysis and regular releasing	Qi et al. [83]
Lutein	NLCs	Protecting from environmental stresses	Mitri et al. [84]
Resveratrol	Peanut oil-based and sunflower oil-based nanoemulsions	Protecting resveratrol from chemical changes	Sessa et al. [67]
Resveratrol	Nanoemulsions by Tween 80	Protecting against degradation	Davidov-Pardo and Mcclements [127]
Resveratrol	Nanoliposomes (cholesterol and diacetyl phosphate)	Enhancing antioxidant activity	Kristl et al. [128]
Resveratrol	SLNs	Enhancing half-life to achieve the therapeutic concentration at the site of action	Jose et al. [129]
Piperine-rich black pepper extracts	Soy phosphatidyl choline/Tween 80 nanoliposomes	Higher antioxidant potency and better storage stability	Dutta and Bhattacharjee [76]
Astaxanthin	SLNs (stearic acid, glycerin monostearate, and glycerol stearates(	Enhancing bioavailability, water solubility, and stability against light, heat, and oxygen	[130]
Vitamin E (α-tocopherol)	Nanoemulsions (lauric acid, myristic acid, palmitic acid, and stearic acid (solid lipid); sunflower oil and olive oil (liquid oil))	Sustaining controlled release and increasing antioxidant activity	Pinto et al. [131]
β-Sitosterol	NLCs (Precirol (lipid), Miglyol(oil))	Increasing the antioxidant activity and storage stability	Bagherpour et al. [132]
Olive leaf phenolics	Nanoliposomes	Improving the antioxidant properties	Tavakoli et al. [133]
Luteolin	Nanoliposomes	Enhancement of solubility and bioavailability	Wu et al. [134]
Cyanidin-3-glucoside	Nanoliposomes	Improving antioxidant activity and bioavailability	Liang et al. [135]
Green tea extract	Nanoliposomes	Improving antioxidant activity and storage stability	Naghavi et al. [136]
α-tocopherol	Niosomal whey protein cold-set hydrogels	A novel functional food formulation and superb delivery potential to preserve niosomes from adverse environmental and/or gastrointestinal conditions	Abaee and Madadlou [86]
Quercetin	Niosomes	Improving solubility, stability, and antioxidant activity retention	Lu et al. [87]
Gallic acid, curcumin	Niosomes	Improving stability, and antioxidant activity retention	Tavano et al. [88]
EGCG	Niosomes	Antioxidant stability	Isnan and Jufri [89]
EGCG	Niosomes	Improving the antioxidant activity in the gastrointestinal tract	Liang et al. [90]
Curcumin	Hexosome	Sustained-release and antioxidant stability	Rakotoarisoa and Angelova [91]

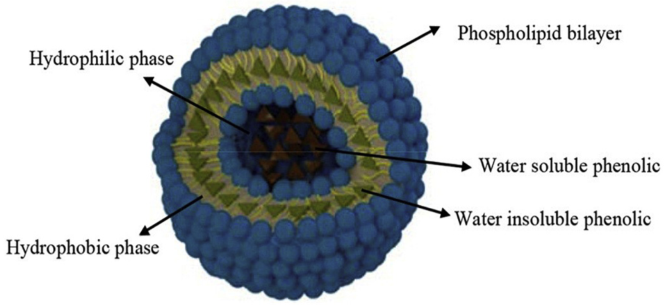

Fig. 3. Illustration of nanoencapsulated phenolics within nanoliposomes (<200 nm); reprinted with permission from [13].

Lu et al. [87] designed flavonoid-loaded niosomes to improve its solubility, stability, and penetration. *In vitro* anti-tyrosinase and DPPH free radical scavenging experiments showed that quercetin has considerable whitening and antioxidant capacities and Span60-RH40 at a mass ratio of 9:11 formed spherical or oval niosomes of 97.6 ± 3.1 nm size and entrapment efficiency as high as 87.3 ± 1.6%. Niosomes remarkably improved the solubility and photostability of quercetin. Furthermore, compared to quercetin solution, quercetin-niosomes had the advantages of sustained release and improved transdermal penetration, with antioxidant activity retention 2.95 times higher than free quercetin solution. Tavano et al. [88] designed niosomal formulations containing antioxidants such as gallic acid, curcumin and in combination, to evaluate the effect of their co-encapsulation on the physicochemical properties of the carriers, their antioxidant activity and capability of releasing the encapsulated materials. Results suggested that the co-encapsulations of gallic acid/curcumin influenced their physicochemical properties and entrapment efficiencies compared with the formulations containing the single antioxidant; also the release of antioxidants appeared to improve and their combinations resulted in a promoted ability of reducing free radicals, due to a synergic antioxidant action; the maximum radical scavenging of gallic acid and curcumin was 60% and 20% respectively, while that of gallic acid/ curcumin was 80%. These amounts were not changed significantly after encapsulation within niosomes.

Isnan and Jufri [89] prepared a topical antioxidant (epigallocatechin gallate; EGCG) preparation based on niosomes. To enhance antioxidant stability, niosomal formulations were prepared in four different molar ratios of surfactant-to-cholesterol, that was, 3:1 (F1), 2:1 (F2), 1:1 (F3), and 0.5:1 (F4) prepared using the thin-layer method. The niosomal suspensions were evaluated for physicochemical properties and encapsulation efficiency, and were then incorporated into gels using hydroxypropyl methylcellulose as the gelling agent. Results showed that F1 had the best encapsulation efficiency but experienced separation after 7 days. The niosomal gels (using F3) showed stable formulation without changes. Result of DPPH radical scavenging activity showed that the EGCG solution had an IC50 of 9.18 ppm; in the niosomal preparation and niosomal gel forms, the IC50 was slightly decreased at 11.97 and 12.51 ppm, respectively. Liang et al. [90] improved the antioxidant activity of EGCG in the GIT. For this purpose, niosomes composed of Tween-60 and cholesterol were developed to encapsulate EGCG, which showed encapsulation efficiency around 76%. The results from ferric reducing antioxidant power and cellular antioxidant activity tests indicated that EGCG-loaded niosomes exhibited stronger antioxidant ability than free EGCG during intestinal digestion. Based on these results it can be concluded that niosomal encapsulation might be a promising approach to improve the oral bioavailability of antioxidants such as EGCG in the body.

The nanoencapsulation of thyme essential oils in nano-niosomes was performed by Emtiazi et al. [174]. The encapsulation efficiency of essential oil was 17% when appropriate formulation was used. They observed that total phenolic contents and antioxidant activity of essential oils was significantly increased after encapsulation. The total phenol content of free and nano-niosomal essential oils were 295.4 and 1523 mg gallic acid g^{-1} essential oils, respectively. Antioxidant capacity for free and nano-niosomal essential oils were 1.13 and 4.2 mmol Fe^{2+} g^{-1} essential oils, respectively. Nanoencapsulation effectively enhanced the beneficial properties of thyme essential oils such as total phenolic contents and antioxidant properties.

A recent study of an inverse hexagonal (HII) liquid crystalline phase encapsulating curcumin by Rakotoarisoa and Angelova [91], demonstrated that the release of curcumin was a concentration-diffusion controlled process in the early stages, whereas multiple diffusion mechanisms coexisted in the later stages of antioxidant release. Radical scavenging experiments showed that curcumin-loaded LCNP[3]s exert the desired antioxidant activity. Thus, curcumin-loaded LCNPs may be promising for neurodegenerative disease treatments using sustained-release nanoformulations for combination therapies.

2.3. Nanocarriers made with specially designed equipment

Several innovative instruments (i.e. electrospinning, electrospraying and nano spray dryer) have been employed to nanoencapsulate antioxidants. Table 3 presents a summary of nanocarriers made with specially designed equipment loaded with antioxidant compounds. Electrospinning can be employed to load phenolic compounds and antioxidants in the nanofibers [21]. For this purpose, biopolymers including proteins (e.g., WPI or WPC, casein, and zein) and carbohydrates (e.g., starch, pectin, cellulose, gums, and chitosan) have exhibited promising potential as the nanoencapsulation systems for antioxidants [92]. Schematic of nanoencapsulated phenolics by specially designed equipment is presented in Fig. 4.

Electrospinning was successfully applied to incorporate quercetin and ferulic acid into amaranth protein isolate and pullulan ultrathin fibers with sustained *in vitro* release compared to the free samples [93]. Neo et al. [94] fabricated gallic acid-loaded zein ultra-fine fibers through electrospinning. The encapsulated gallic acid maintained its phenolic nature and antioxidant properties after electrospinning. Ranjbar-Mohammadi and Bahrami [95] employed electrospinning to prepare poly (ε-caprolactone)/gum tragacanth/curcumin-loaded nanofibers. They optimized electrospinning parameters (feed rate, voltage, nozzle-collector distance and polymer concentration) to obtain fibers with a minimal diameter through response surface methodology (RSM) and artificial neural networks (ANNs). The results were indicative of ANNs and RSM models consistency with the predicted fiber diameter. According to RSM, minimum diameter was achievable using polymer at concentration of 4.2% (w/v), with nozzle-collector distance of 20 cm, voltage of 20 kV, and feed rate of 0.5 mL/h. Loading 3% (w/v) curcumin into gum tragacanth/PVA[4] nanofibers resulted in enhanced antioxidant activity introducing it a promising option for disease treatment applications.

Shekarforoush et al. [96] obtained highly stable nanofibers from electrospun xanthan–chitosan viscoelastic gels which were employed to encapsulate curcumin. Owing to its hydrophobic features, curcumin declined the adhesion properties of produced nanofibers. After 120 h, at pH=2.2 and neutral media, curcumin contents were measured as 20% and 50%, respectively. In this regard, due to high encapsulation efficiency, physicochemical stability in aqueous environments and long-term pH-stimulated release, electrospun xanthan–chitosan nanofibers can be recognized as a promising carrier for encapsulating hydrophobic antioxidants. Rezaei and Nasirpour [97] reported production of composite nanofibers consisting of curcumin. The almond gum/PVA/β-cyclodextrin composite showed smaller diameter than almond gum/PVA fiber. Moreover, these nanofibers exhibited improved thermal stability

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[3] - Liquid crystalline nanoparticles

[4] - Polyvinyl alcohol

Table 3
Selected studies on nanocarriers made with specially designed equipment for antioxidant compounds.

Antioxidants	Nanocarriers	Special equipment	Purpose	Reference
Curcumin	Chitosan/poly (lactic acid) nanofibers	Electrospinning	Improving the bioavailability	Dhurai et al. [137]
Folic acid	Nanoparticles of whey protein concentrate matrix and a commercial resistant starch	Nano-spray dryer	Physicochemical stability and preventing degradation	Perez-Masia et al. [171]
Quercetin and ferulic acid	Amaranth protein isolate and pullulan nanofibers	Electrospinning	Sustained through *in vitro* digestion and improving antioxidant capacity	Aceituno-Medina et al. [93]
Superoxide dismutase	Poly (lactic acid) nanofibers	Electrospinning	Controlled release system	Chen et al. [138]
Curcumin	Poly (ε caprolactone)/gum tragacanth nanofibers	Electrospinning	Increasing antioxidant activity	Ranjbar-Mohammadi and Bahrami [95]
Gallic acid	Zein nanofibers	Electrospinning	Preserving antioxidant activity	Neo et al. [94]
Crocins and picrocrocin	Maltodextrin	Nano-spray dryer	Increasing the thermal stability and bioaccessibility	Kyriakoudi and Tsimidou [172]

upon curcumin incorporation. The almond gum nanofiber managed to enhance the curcumin solubility; thus it can be regarded as a promising carrier to preserve the hydrophobic antioxidants.

Kyriakoudi and Tsimidou [172] studied the production, characterization and stability of saffron hydrophilic apocarotenoids, i.e. crocins and picrocrocin, in maltodextrin using nano spray-drying. The effect of mesh size and core:wall ratio (w/w) on the product yield and encapsulation efficiency of these apocarotenoids was examined. Final nanoparticles were characterized and their stability was examined under thermal and *in vitro* gastrointestinal conditions. Spherical particles were obtained. Product yield and encapsulation efficiency (%) of crocins and picrocrocin was found to be satisfactory. Thermal stability and bioaccessibility of these apocarotenoids was also enhanced by nanoencapsulation.

2.4. Nature-inspired nanocarriers

Nature-inspired nanocarriers have drawn a considerable deal of attention in recent years since they can serve as safe and cost-effective tools of drugs and nutrients delivery. The features of these natural nanocarriers have been classified according to their biological source, isolation methods and physical and biochemical alterations. These miniature natural NPs (i.e. caseins, cyclodextrins, amylose and starch granules) can be exploited for nanoencapsulation of antioxidants [16]. Table 4 briefly describes antioxidant-loaded natural nanocarriers.

The physical and chemical characteristics of casein are similar to copolymers; it possesses well-balanced hydrophobic and hydrophilic zones capable of self-assembling into nano-scaled carriers. Moreover, its superior thermal stability has introduced it as a promising natural carrier for sensitive antioxidants. Owing to their structural and physicochemical features combined with their tendency for hydrophobic interactions, caseinates and β-caseins have been widely employed for encapsulation of various hydrophobic antioxidants including β-carotene and curcumin [16]. The hydrophobic interactions occurring between curcumin and β-casein micelles may enhance the curcumin solubility, bioavailability and antioxidant properties [98]. The particle size of native casein micelles can reach to 500 nm; it could be however reassembled to finer sizes [99]. Different approaches can be employed to load β-carotene into reassembled casein micelles such as high-pressure homogenization or microfluidization. Both methods provide high dissociation energy for casein micelles [100].

Due to its open structures with several evenly-distributed proline residues over its amino acids, casein has been considered as a promising binder for phenolic compounds and antioxidants. The hydrophobic interactions between casein micelles and β-carotene can elevate the β-carotene bioavailability, stability and solubility. Semo et al. [99] loaded β-carotene into casein micelles and evaluated them under several industrial processes such as heating, sterilization, pasteurization, high hydrostatic pressure and baking. Compared to free β-carotene, casein micelles were able to protect β-carotene from degradation during all the mentioned conditions. In another study, Yi et al. [101] employed emulsification-evaporation lyophilization technique to nanoencapsulate β-carotene in casein. Tan and Nakajima [102] and Yi et al. [101] indicated that sodium caseinate nanocarriers possessed the smallest particle size, compared with whey protein and soy protein isolate. The medium effective dose value of β-carotene against Caco-2 cells in nanoencapsulated form was about half of the free β-carotene [101].

Esmaili et al. [98] applied β-casein for curcumin encapsulation through solvent evaporation technique. The encapsulated curcumin exhibited stronger antioxidant behavior as compared with free curcumin. The medium effective dose of encapsulated curcumin on human leukemia cell line K-562 was decreased to 17.7μmol/L. Casein-zein complex, casein-zein-pectin complex [103], casein-dextran conjugate [104], and

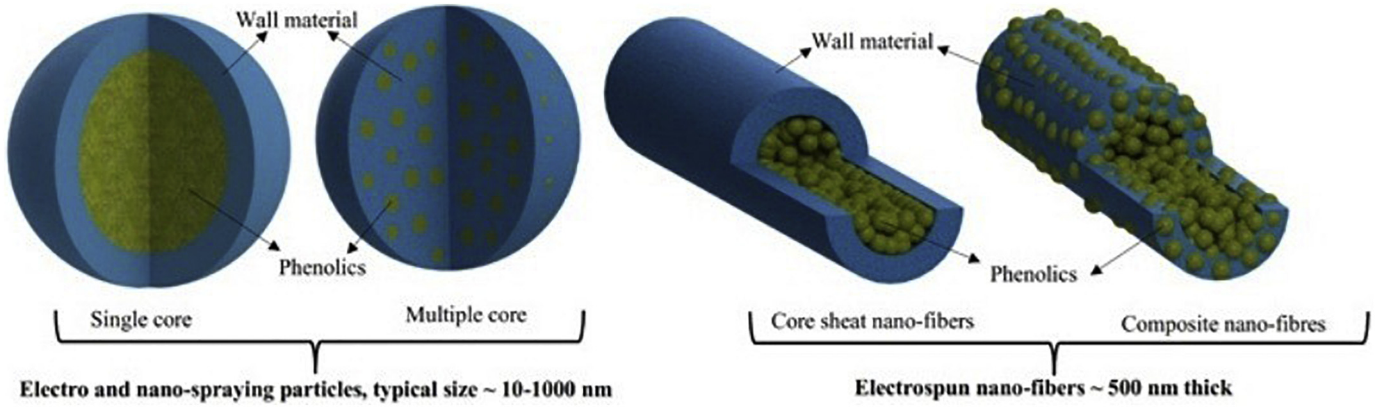

Fig. 4. Schematic representation of phenolics-loaded nanocarriers prepared by specially designed equipment; reprinted with permission from [116].

Table 4
Some studies on nature-inspired nanocarriers loaded with antioxidant compounds.

Antioxidants	Nanocarriers	Purpose	Reference
β-carotene	Casein micelles	Increasing the bioavailability, stability and solubility of β-carotene	Chu et al. [100].
β-carotene	Casein micelles	Protecting the degradation of β-carotene	Semo et al. [99].
β-carotene	Casein micelles	Decreasing the medium effective dose values of β-carotene against Caco-2 cells	Yi et al. [101]
Curcumin	β-casein micelles	Increasing the antioxidant activity and decreasing the medium effective dose of curcumin tested on human leukemia cell line K-562	Esmaili et al. [98]
Curcumin	Casein-soy polysaccharides	Maintaining antioxidant activity, extending storage stability and controlling release	Wu and Wang [104]; Xu et al. [105]
Curcumin	Casein-zein complex, casein-zein-pectin complex	Maintaining antioxidant activity and extending storage stability	Chang et al. [103]
Curcumin	Casein-dextran conjugate	Maintaining antioxidant activity, extending storage stability and improving oral bioavailability	Wu and Wang [104]
Curcumin	Cyclodextrins	Increasing water solubility and maintaining antioxidant activity	Yallapu et al. [108]
Curcumin	β-casein micelles	Increasing the solubility of curcumin and its bioavailability and antioxidant activity	Esmaili et al. [98]
Rutin, tannic acid, catechin, and gallic acid	Casein based films	Producing radical-scavenging films, applicable to food products that are susceptible to oxidation	Helal et al. [106]
Usnic acid	β-cyclodextrin incorporated into liposomes	Providing a targeted delivery system	Lira et al. [109]
Catechins	β-cyclodextrin	Increasing antioxidant activity	Krishnaswamy et al. [110]
Catechins	β-casein micelles	Increasing water solubility and maintaining antioxidant activity	Haratifar and Corredig [139]
Resveratrol	Cyclodextrins	Increasing bioavailability and stability	Lucas-Abellan et al. [111]
Oleoresin	Cyclodextrins	Enhancing the antioxidant activity	Teixeira et al. [140]
Astaxanthin	Cyclodextrins	Increasing stability and water solubility	Yuan et al. [141]
Naringenin	β-casein micelles	Enhancing solubility and stability	Moeiniafshari et al. [142]

casein-soy polysaccharide [105] are among the encapsulating biopolymers used for curcumin loading. Encapsulated curcumin has shown enhanced storage stability [104,105] and controlled release [103,105] in simulated digestive juices, as well as augmented oral bioavailability [105]. Other strong antioxidant polyphenols (e.g. rutin, tannic acid, catechin, and gallic acid) could be incorporated in NaCas-based films to fabricate radical-scavenging films with possible application in oxidation-susceptible food products [106].

Cyclodextrins (CDs) refer to a class of hollow molecular nanocarriers with specific sizes which can be used in encapsulation of various antioxidants by encompassing an appropriate "guest" structure into their cavity [107]. Hydrophobic components of CDs may surround the phenolics and antioxidants and protect them against the environmental factors (such as pH, light, oxygen, and temperature); while improving their water solubility. Schematic of nanoencapsulated phenolics by β-CD and casein micelles is shown in Fig. 5.

To mention an example, curcumin was loaded inside the hydrophobic zone of CDs to increase its water solubility and maintain its antioxidant properties [108]. Lira et al. [109] used inclusion complexation and freeze drying procedures to encapsulate usnic acid (UA) (an antioxidant) into β-CDs and form a complex (UA/β-CD). The resulted complex (UA/β-CD) was further added to liposomes to develop a targeted delivery system. UA/βCD-loaded liposomes were also synthesized through hydration of a thin lipid film followed by sonication. These liposomes sustained their stability for 4 months without any alternation in their antioxidant activity. Interesting result was that UA/βCD encapsulation into liposomes led to modulated *in vitro* release kinetics of UA. UA/β-CD-loaded liposomes exhibited a prolonged usnic acid release profile in comparison with UA-loaded liposomes [109]. Yallapu et al. [108] reported that in comparison with free curcumin, CD-encapsulated curcumin showed improved delivery and antioxidant properties as well as enhanced therapeutic efficacy toward prostate cancer cells. In another study, catechins were successfully nanoencapsulated into β-CD at the ratio of 1:1 through molecular inclusion method which led to higher antioxidant activity of catechins [110]. CDs have also served as a carrier to improve the resveratrol bioavailability and stability by forming inclusion complexes [111].

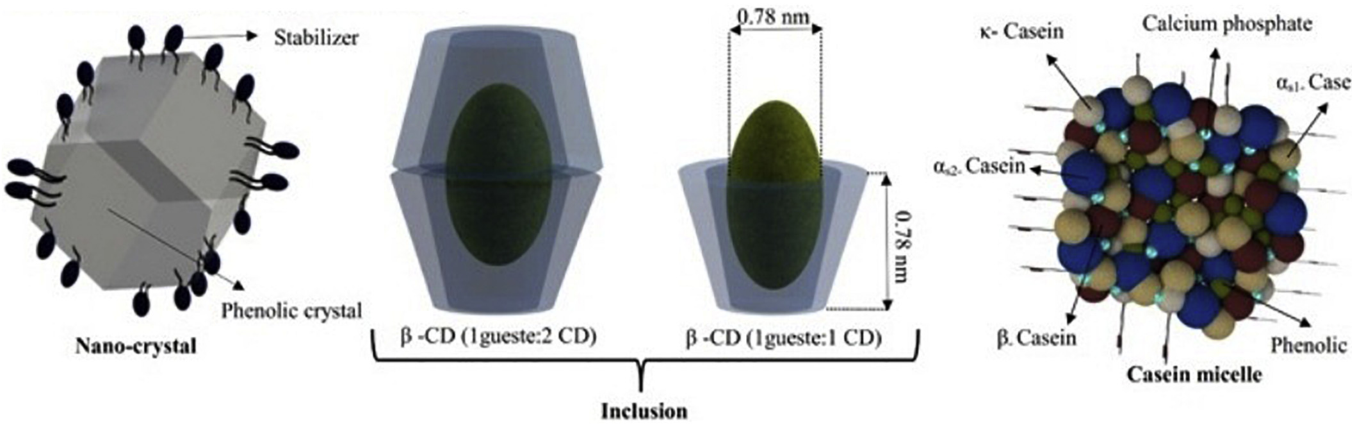

Fig. 5. Illustration of nanoencapsulated phenolics within nature-inspired nanocarriers; reprinted with permission from [116].

Table 5
Some studies on the application of antioxidant-loaded nanocarriers in real food products.

Antioxidants	Nanocarriers	Food system	Results	References
Combination of astaxanthin, α-tocopherol and EDTA	NLCs	Non-pasteurized CO_2-free beer	Improved stability at low storage temperature of 6 °C	Tamjidi et al. [143]
Quercetin	Nanoemulsions	Beverage formulations	No negative effects on the texture and appearance of the beverage during storage	Ni et al. [144]
Hydroxytyrosol, an antioxidant of olive oil	Double emulsion solvent evaporation	Foods, drugs, and nutraceuticals	Antioxidant capacity preservation	Mokhtari et al. [48]
β-carotene	Nanoemulsions	Food supplements	Better stability against droplet aggregation under simulated GIT conditions (mouth, stomach, and small intestine)	Lin et al. [145]
Epigallocatechin gallate (EGCG) and catechin	Nanoliposomes	Ripened low-fat cheese	Increased antioxidant activity of cheese after simulated GIT digestion without any notable effects on pH, chemical composition, and production yield. Moreover, EGCG and catechin completely remained in cheese structure and were not detected in whey	Rashidinejad et al. [146]
Flavonoids from extract of Tartary buckwheat	Biocompatible lipid–polymer hybrid nanoparticles	Nutraceutical supplements	Higher antioxidant activity in immunosuppressed mice	Zhang et al. [147]
Rutin	NLCs (oleic acid, cacao butter, and Tween)	Beverages (orange juice, milk, apple juice)	Fortified beverages, were physically and thermally stable and showed an acceptable sensorial quality	Babazadeh et al. [148]
Rutin	Hexosome	Beverages (orange juice, milk, apple juice)	Stable structure and acceptable sensorial quality of beverages	Babazadeh et al. [148]
Vitamin C	Nanoliposomes	Mandarin juice	Higher stability of the fortified mandarin juice with a negligible effect on the sensory properties of juice	Liu et al. [149]
β-sitosterol	NLCs	Butter	No evident change at peroxide and acid values of butter samples containing NLCs; good oxidative stability and maintained antioxidant activity of loaded β-sitosterol in butter matrix over 3 months of storage at 4^0C	Bagherpour et al. [150]
α-tocopherol	Low-molecular-weight octenyl succinic anhydride modified starch nanocarriers	Supplemental food and beverages	Better stability	Hategekirnana et al. [151]
Curcumin and resveratrol	Liposomes	Nutritional supplements	Excellent stability and sustained release character and higher radical scavenging activity	Guo et al. [152]
Carvacrol and Thymol	β-cyclodextrin nanoparticles	Ice (for storage of fresh whole fish in ice)	Improved the quality of fresh fish and extended the shelf-life up to 4 days	Navarro-Segura et al. [153]
Catechins	Chitosan nanoparticles	Gelatin films with controlled-release properties for fatty food simulants	Films maintained their structures and antioxidant activity after 240 h	Liu et al. [154]
Olive leaf extract	Nanoemulsions	Soybean oil	Maintaining the oxidative stability of soybean oil	Mohammadi et al. [155]
Green tea extract, chlorogenic acid	Chitosan nanoparticles	An edible coating for walnut kernels	The best result at reducing the oxidation activity and undesirable sensory properties	Sabaghi et al. [156]
Coffee hydroxycinnamic acids (HCAs)	β-cyclodextrins	Cookies, bread, caramel cottage, mushroom and meat stuffing and nutty filling	Protection and stability of HCAs during the processing	Budryn et al. [157]
Green tea extract (chlorogenic acid and other polyphenols)	Nanoparticles of maltodextrin, β-cyclodextrin and their combination	Bread	Bread incorporated with nanocarriers showed better color and taste than the one with free green tea extract. Although bread characteristics such as volume, crumb firmness and total polyphenols content seemed to be similar to control	Pasrija et al. [158]
Myristica fragrans essential oil	Chitosan nano-matrix	Rice seeds during storage	Promising inhibitory action on lipid peroxidation in stored rice seeds	Das et al. [159]
α- tocopherol	Amylose from corn starch as complexes with flax seed oil	Functional bread	Increased final product quality and safety by lowering lipid oxidation and lower formation of harmful compounds in breads during baking	Gökmen et al. [160]
Rosmarinus officinalis essential oils	Nanogels	Beef cutlet	Improved antioxidant activity in cutlets without any changes in the sensory properties	Hadian et al. [161]
Lyophilized pomegranate peel	Nanoparticles of chitosan	Meatballs of minced beef meat	Prevented lipid oxidation and improved cooking characteristics; the shelf-life of meatballs was extended up to 15 days, without any negative impact on sensory properties of both raw and cooked meatballs	Morsy et al. [162]
Thyme essential oil	Nanoparticles of chitosan	Beef burgers	Control of undesirable lipid oxidation, and sensory changes	Ghaderi-Ghahfarokhi et al. [163]
Jabuticaba extract	Maltodextrin nano-particle	Fresh sausage	Reduction in lipid oxidation in fresh sausage	Baldin et al. [164]
Cinnamon essential oil	Chitosan nanoparticles	Beef patties	Good preservative properties, strong flavor of cinnamon essential oil had adverse effect on red color	Ghaderi-Ghahfarokhi et al. [163]
Clove essential oil	Chitosan-myristic acid nanogels	Beef cutlets	Reduction in lipid oxidation	Hadian et al. [161]
β-carotene	Nanoparticles cross-linked chitosan with sodium tripolyphosphat	Hamburger patties	Reduction in lipid oxidation	Ozvural and Huang [165]

(continued on next page)

Table 5 (*continued*)

Antioxidants	Nanocarriers	Food system	Results	References
Curcumin	Polyvinyl pirrolidone nanoparticles	Yogurt	The best result at reducing the oxidation activity and undesirable sensory properties	Almeida et al. [166]
Green tea catechins and EGCG	Liposomes	Hard low-fat cheese	Catechins and EGCG were not detected in the cheese whey, indicating complete retention in the cheese structure; increasing the antioxidative functionality of cheese by releasing under GIT digestive conditions	Rashidinejad et al. [167]
Turmeric extract (including curcumin)	Nanoemulsions	Milk	Good preservative properties; aroma of milk was not noticeably affected	Park et al. [168]

3. Application of antioxidant-loaded nanocarriers in real food products

Fortification of food products with bioactive compounds such as antioxidants has attracted growing consideration due to the increased awareness and tendency of consumers towards healthy diets (Faridi [21]). Enrichment/fortification of foods by antioxidant compounds not only improves their stability, but also can be effective in prevention and cure of some diseases such as cardiovascular and neurodegenerative diseases [112]. Bioactive antioxidants are suitable to be used for the fortification of various food products to enhance their functionality, and therefore, encapsulation systems for the delivery of such nutraceuticals are necessary to overcome their low stability and bioavailability [113]. The choice of the appropriate encapsulation method is essential, because the modification of bioactivity (increase, preservation, or decrease) is affected by interactions established between the functional groups of the encapsulated compound and the encapsulating nanomaterial [114]. In the development and optimization of a product, regulatory issues should also be taken into account, along with the cost of production and the preferences of the consumer. Efforts have been made towards loading antioxidant molecules in advanced nanoparticles, such as SLNs, NLCs, polymeric nanoparticles, liposomes and emulsions [115]. Table 5 presents a summary of the applied antioxidant-loaded nanocarriers in real food products.

4. Conclusion

Nanoemulsions, nanoliposomes, nanostructured lipid carriers and solid lipid nanoparticles are the major studied and developed types of lipid-based and surfactant-based nanocarriers in the recent years. The loaded antioxidants showed enhanced stability and bioavailability in comparison with non-encapsulated ones. Several innovative instruments (i.e. electrospinning, electrospraying and nano spray dryer) have also been employed to nanoencapsulate antioxidants. The encapsulated antioxidants maintained their nature and antioxidant properties after electrospinning. Nature-inspired nanocarriers have drawn a considerable deal of attention in recent years since they can serve as safe and cost-effective tools for antioxidant delivery. The physical and chemical characteristics of casein are similar to copolymers; it possesses well-balanced hydrophobic and hydrophilic zones capable of self-assembling into nano-scaled carriers. Moreover, its superior thermal stability has introduced it as a promising natural carrier for sensitive antioxidants. Owing to their structural and physicochemical features combined with their tendency for hydrophobic interactions, caseinates and β-caseins have been widely employed for encapsulation of various hydrophobic antioxidants. Cyclodextrin refers to a class of hollow molecular nanocarriers with specific sizes which can be used in encapsulation of antioxidants by encompassing an appropriate “guest” structure into their cavity. Hydrophobic components of cyclodextrins may surround the phenolics and antioxidants and protect them against the environmental factors while improving their water solubility. Despite the minor negative effects of nanoencapsulation, this method largely retains the antioxidant properties of bioactive compounds. Even in some cases, due to the type of wall material, it may increase the antioxidant activity of the encapsulated compounds. It has been verified that antioxidant-loaded nanocarriers can be applied in many formulations with a higher and controlled release antioxidant activity.

Declaration of Competing Interest

All authors declare that there is no conflict of interest.

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