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Review

Nanoencapsulation of food ingredients using carbohydrate based delivery systems

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Natural and modified polysaccharides are promising vehicles for nano- and micro-encapsulation of active food ingredients. This article reviews the state of the art of carbohydrate-based delivery systems for utilization in the food, pharmaceutical and other industries. Initially, an overview of the different kinds of carbohydrates used to assemble delivery systems is given, including starch, cellulose, pectin, guar gum, chitosan, alginate, dextrin, cyclodextrins, new sources of native gums, and their combinations and chemically modified forms. Their molecular and physicochemical properties, functional performance, and advantages and disadvantages for encapsulation are given. Various approaches for fabrication of carbohydrate-based delivery systems are then discussed, including coacervation, spray drying, electrospinning, electrospray, supercritical fluid, emulsion–diffusion, reverse micelle, emulsion-droplet coalescence, emulsification/solvent evaporation, salting-out, ultrasonication and high pressure homogenization. The biological fate of carbohydrate nanocarriers during digestion, absorption, metabolism and excretion are discussed, and some notes about their bioavailability and potential toxicity are provided. Finally, the functional performances of different carbohydrate-based delivery systems are discussed, and future developments are highlighted.

Introduction

Nanotechnology is defined as creation, utilization and manipulation of materials, devices or systems in the nanometer scale (smaller than 100 nm). Nanoencapsulation as an important field of nanotechnology is a process that involves entrapping bioactive agents within carrier materials with a dimension in nano-scale. Enormous demands for production of functional food with higher nutritional value, lower dose of synthetic preservatives and better organoleptic features lead to innumerable applications of nanoencapsulation in food processing. For example this technology has be used to enhance the stability of sensitive compounds during production, storage and ingestion, *e.g.* vitamins (Khayata, Abdelwahed, Chehna, Charcosset, & Fessi, 2012; Sáiz-Abajo, González-Ferrero, Moreno-Ruiz, Romo-Hualde, & González-Navarro, 2013), decrease evaporation and degradation of volatile bioactives, *e.g.* aromas (Donsì, Annunziata, Sessa, & Ferrari, 2011), mask unpleasant tastes, *e.g.* polyphenols (Fathi, Varshosaz, Mohebbi, & Shahidi, 2013), or limit exposure to oxygen, water or light, *e.g.* unsaturated fatty acids (Nedovic, Kalusevic, Manojlovic, Levic, & Bugarski, 2011; Zimet & Livney, 2009). The encapsulating carrier material must be food-grade, biodegradable, and stable in food systems during processing, storage, and consumption. The most suitable nano-scale carrier materials for food applications are carbohydrate-, protein- or lipid-based. In previous works the state of knowledge of lipid-based delivery systems were discussed (Fathi, Mozafari, & Mohebbi, 2012; McClements & Li, 2010). Polysaccharide-based delivery systems are suitable for many industry applications since they are biocompatible, biodegradable, and possess a high potential to be modified to achieve the required properties. In contrary to the lipid carriers, carbohydrate based delivery systems can interact with a wide range of bioactive compounds

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via their functional groups, which makes them versatile carriers to bind and entrap a verity of hydrophilic and hydrophobic bioactive food ingredients. On the other hand, they are considered as a suitable shell under high temperature processes due to their temperature stability in comparison to lipid or protein based delivery systems which might be melted or denatured. This review critically assesses different carbohydrates and assembly techniques that can be used to fabricate delivery systems, and highlights some of the advantages and disadvantages of different systems. The biological fate of carbohydrate nanocarriers during digestion, absorption, metabolism and excretion are discussed, and some notes about their bioavailability and potential toxicity are provided. Finally, potential future developments in this rapidly growing area are highlighted.

Carbohydrate building blocks

In this section, we review the various types of natural and modified carbohydrates that can be used to create nano-scale encapsulation and delivery systems. Carbohydrates consist of monosaccharides, oligosaccharides, and polysaccharides. However, due to their massive molecular structure and therefore ability to entrap bioactives the later are most suitable as building blocks of delivery systems. Polysaccharides are natural polymers of monosaccharides that vary in the type, number, distribution, and bonding of the monomers in the chain (Bochkov, Zaikov, & Afanas'ev, 1991). They have attracted considerable attention for delivery of bioactive food components because they are already widely used as safe and inexpensive food ingredients, whose functional attributes are tailored by their biological origins, isolation methods, or physical, chemical or biochemical modifications. Generally carbohydrate based delivery systems are conveniently categorized according to their biological origins: plant origin (*e.g.* starch, cellulose, pectin, and guar gum); animal origin (*e.g.* chitosan); algal origin (*e.g.* alginate and carrageenan), and microbial origin (*e.g.* xanthan, dextran and cyclodextrins) (Eliasson, 2006). In the following sections, different molecular and physicochemical properties of the carbohydrate based delivery systems are reviewed.

Starch

Starch, which is the most abundant storage polysaccharide in plants, is composed of glucose units linked with α-d-(1 → 4) and/or α-d-(1 → 6) linkages. It is a biodegradable, biocompatible, and digestible polymer that consists of two main structural components: amylose and amylopectin (Barsby, Donald, Frazier, & Royal Society of Chemistry. Food Chemistry, 2001). Starch based-nanoparticles have been used for encapsulation of insulin (Jain, Khar, Ahmed, & Diwan, 2008), flax seed (Gökmen *et al.*, 2011), unsaturated fatty acids (Lesmes, Cohen, Shener, & Shimoni, 2009; Zabar, Lesmes, Katz, Shimoni, & Bianco-Peled, 2009), and flavors (Itthisoponkul, Mitchell, Taylor, & Farhat, 2007). Natural starch is predominantly hydrophilic, which limits its application for encapsulating hydrophobic food bioactives. Hydrophobic starch derivatives have therefore been developed to overcome this drawback. Dialdehyde starch (Yu, Xiao, Tong, Chen, & Liu, 2007) and propyl starch (Jain *et al.*, 2011; Santander-Ortega *et al.*, 2010) nanoparticles have been fabricated for encapsulation and delivery of lipophilic pharmaceutical agents. Octenyl Succinic Anhydride (OSA) modified starches have also been used to encapsulate food and flavor ingredients (Qi & Xu, 1999). OSA-starches are surface active molecules that absorb to oil—water interfaces and facilitate the formation of small oil droplets during homogenization, and increase their long term stability during storage. Acetylation of starch has been shown to increase its hydrophobicity, reduce swelling ability, and enhance resistance to enzymatic hydrolysis, which are useful attributes for development of delivery systems for pharmaceutical agents (Tuovinen *et al.*, 2004; Xu, Yang, & Yang, 2009) and food ingredients (Pu *et al.*, 2011). PEGylated starch acetate nanoparticles, which have a hydrophobic core, have been used for oral insulin delivery, and showed higher encapsulation efficiency than starch acetate nanoparticles (Minimol, Paul, & Sharma, 2013). Some more examples of modified starch and other carbohydrates are shown in Table 1.

Linear amylose molecules are known to co-crystallize with various lipophilic molecules, *e.g.* long-chain alcohols, aromatic compounds, lipids and surfactants (Zabar *et al.*, 2009). The amylose molecules form helices with a hydrophobic cavity that can trap water-insoluble food bioactives with particular molecular geometries. Recent studies have shown that the structure, physicochemical properties, amylase digestibility, and release behavior of these amylose-complexes depends upon the chemistry of the guest molecules (Gökmen *et al.*, 2011).

It is noteworthy that apart from the low cost of starch, it is relatively pure and does not need intensive purification procedures like other biopolymers, such as cellulose and some gums. The main limitation of starch application arises from its sensitivity to acid attack and amylase hydrolysis which might be started in mouth; however, as mentioned above some modifications could be applied for production of acid- or enzyme-resistance starch.

Cellulose

Cellulose, which is the most abundant polysaccharide on earth, is composed of glucose units linked with β-d-(1 → 4) linkages. In its natural form it is not particularly suitable as a building block of delivery systems because of its low-water solubility and large dimensions. Nevertheless, this biopolymer can be physically, chemically, or biochemically modified to produce more suitable building blocks as encapsulating shell. Nanocrystal cellulose (NCC) was first proposed by Favier *et al.* (1995) and is produced by acid hydrolysis of cellulose from sources such as linter (Lin, Huang, Chang, Feng, & Yu, 2011). NCC has

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