1	Food Hydrocolloids: Application as Functional Ingredients to Control
2	Lipid Digestion and Bioavailability
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29 Abstract

30 One of the most popular research areas in food hydrocolloids over the past decade or 31 so has been their application as functional ingredients to modulate the gastrointestinal 32 fate of foods. In particular, they are being utilized to control the hydrolysis of 33 macronutrients (such as fat, protein, and starch) in the gastrointestinal tract, as well as to 34 alter the pharmacokinetics and bioavailability of hydrophobic bioactive agents, including 35 oil-soluble vitamins (e.g., vitamin A, D, E, and K), nutraceuticals (e.g., carotenoids, 36 phytosterols, curcumin, resveratrol, and quercetin) and healthy lipids (e.g., omega-3 fatty 37 acids and conjugated linoleic acids). Food hydrocolloids may be naturally present in 38 foods (such as fruits, vegetables, seeds, and cereals), they may be added as functional 39 ingredients (such as thickening, gelling, emulsifying or stabilizing agents), or used to 40 construct colloidal delivery systems (such as emulsions or microgels). Hydrocolloids can 41 be used to protect bioactive agents from chemical degradation within foods and 42 beverages during storage, but then increase their bioavailability after consumption. 43 Moreover, they can be used to target the delivery of bioactive agents to particular sites in 44 the gastrointestinal tract or to modulate their release profile. Food hydrocolloids are 45 therefore versatile natural ingredients for the formulation of a new generation of 46 functional food products designed to enhance human health and wellbeing. This article 47 provides a review of the application of food hydrocolloids, mainly proteins and 48 polysaccharides, for modulating the gastrointestinal fate of functional foods, with an 49 emphasis on their ability to control macronutrient digestion and bioactive bioavailability. 50 51 Keywords: Dietary fiber; delivery systems; nanoemulsions; microgels; gastrointestinal 52 tract; nutraceuticals

54 **1. Introduction**

55 A major trend in the field of food hydrocolloids over the past decade or so has been 56 the use of proteins and polysaccharides as functional ingredients to modulate the 57 gastrointestinal fate of macronutrients, micronutrients, and nutraceuticals, collectively 58 referred to as "nutrients" in the remainder of this article. These hydrocolloids can be 59 used in a variety of ways to achieve this goal. They can be utilized as building blocks to 60 assemble food-grade colloidal delivery systems to encapsulate, protect, and release 61 nutrients (Shewan & Stokes, 2013; Taheri & Jafari, 2019; Zhang, Zhang, Chen, Tong, & 62 McClements, 2015). They can be used as functional ingredients to alter the nature of the 63 gastrointestinal fluids inside the human body (especially their rheological properties) so 64 as to control the mixing, digestion, and transport of nutrients (Foster & Norton, 2009; 65 Pedersen, et al., 2013; Tharakan, Norton, Fryer, & Bakalis, 2010). They can be used to 66 bind nutrients and gastrointestinal constituents (such as bile acids, enzymes, or calcium 67 ions), thereby altering the digestion and/or absorption of the nutrients within the body 68 (Hu, Li, Decker, & McClements, 2010; Zhou, Xia, Zhang, & Yu, 2006). They can be 69 utilized as prebiotics to change the composition of the gut microflora, thereby improving 70 human health (Wang, et al., 2019). Moreover, the presence of hydrocolloids in natural 71 foods, such as fruits, vegetables, cereals, nuts, or seeds, may be utilized for their ability to 72 modulate the gastrointestinal behavior of foods (Singh & Gallier, 2014). The purpose of 73 this manuscript is to provide an overview of some of the recent research on the utilization 74 of food hydrocolloids to create functional foods and beverages designed to enhance 75 human health, wellbeing, or performance. In particular, the key molecular, 76 physicochemical, and functional properties of hydrocolloids suitable for this purpose are 77 highlighted.

78 2. Food Hydrocolloids

79 There have been many excellent articles and books published on the molecular 80 characteristics, physicochemical properties, and functional performance of food grade 81 hydrocolloids (Li & Nie, 2016; Phillips & Williams, 2009; Stephen, Phillips, & Williams, 82 2006), and readers are referred to these publications for more detailed information about 83 specific hydrocolloids. Briefly, the two most important types of functional hydrocolloids 84 in foods are proteins and polysaccharides (DNA or RNA are rarely used as functional 85 food ingredients). Most food proteins are polymers consisting of a linear chain of amino 86 acids linked together by peptide bonds. The number, type, and sequence of the amino 87 acids determines the molecular weight, conformation, charge, and polarity of the proteins, 88 which in turn impacts their solubility, surface activity, thickening, gelling, foaming, 89 enzyme activity, and nutritional attributes. Some important food proteins have other 90 groups covalent attached to the polypeptide chain, such as casein (phosphates), 91 lactoferrin (sugars), or myoglobin (heme), and hemoglobin (heme), which play a key role 92 in their functionality. Polysaccharides are polymers consisting of monosaccharides 93 linked together by glycosidic bonds. The type, number, sequence, and bonding of the 94 monosaccharides impacts their molecular and functional properties. A number of the 95 most important food hydrocolloids used to modulate the gastrointestinal fate of foods are 96 summarized in Table 1 along with their key molecular and functional attributes.

97 **3. Key Functional Attributes Impacting Gastrointestinal Fate**

98 Food hydrocolloids can modulate the gastrointestinal fate of foods in a variety of 99 ways (Blackwood, Salter, Dettmar, & Chaplin, 2000). In this section, some of the key 100 functional attributes of food hydrocolloids that impact their behavior inside the human 101 gut are briefly discussed.

102 **3.1. Solubility**

103 Food hydrocolloids may be fully soluble, partially soluble, or insoluble in water (or 104 gastrointestinal fluids) depending on their molecular characteristics, particularly their 105 surface hydrophobicity, charge, and molecular weight (Guo, Hu, Wang, & Ai, 2017a). 106 Water solubility tends to decrease with increasing surface hydrophobicity because of the 107 increase in hydrophobic attraction between the non-polar patches on the surfaces of 108 different hydrocolloid molecules. This is the main reason that hydrophobic proteins, such 109 as zein and gliadin, have low water-solubilities (Davidov-Pardo, Joye, & McClements, 110 2015). The water-solubility may also be low if there are relatively strong hydrogen bonds 111 between linear polysaccharide chains that can come close together, as in cellulose and 112 mannan (Guo, et al., 2017a). The water-solubility of hydrocolloids tends to decrease as 113 their electrical charge decreases because of the reduction in electrostatic repulsion

114 between them (Curtis & Lue, 2006). This is the reason why many proteins tend to 115 aggregate and precipitate close to their isoelectric points (Gehring, Gigliotti, Tou, Moritz, 116 & Jaczynski, 2010). Conversely, highly charged hydrocolloids may precipitate in the 117 presence of oppositely charged substances due to an electrostatic attraction between, e.g., 118 when an anionic polysaccharide is mixed with a cationic protein (Weiss, Salminen, Moll, 119 & Schmitt, 2019). Hydrocolloid solubility tends to decrease with increasing molecular 120 weight because of the reduction in the entropy of mixing, which favors the random distribution of the hydrocolloids throughout the solvent (Curtis, et al., 2006). 121

To a first approximation, the solubility of hydrocolloids (especially globular
proteins) can be treated theoretically by assuming they are colloidal particles and
calculating the various colloidal interactions between them, such as van der Waals, steric,
hydrophobic, and electrostatic (Curtis, et al., 2006): if the molecules tend to aggregate
they have a low solubility, but if they tend to stay apart they have a high solubility.

127 The functionality of many hydrocolloids depends on their solubility in the aqueous 128 phase of foods, as well as in the aqueous gastrointestinal fluids. The ability of proteins or 129 polysaccharides to thicken solutions, form gels, or stabilize emulsions often requires that 130 they have a good water-solubility (Phillips, et al., 2009). Thus, hydrocolloids that are 131 used for this purpose, such as gum arabic, modified starch, whey proteins, caseins, and 132 soy proteins should have good water-solubility characteristics (at least under the required 133 solution conditions). Conversely, hydrocolloids that are insoluble in water, such as zein or 134 gliadin, are useful for constructing colloidal particles that can be used as delivery systems 135 in foods (Fathi, Donsi, & McClements, 2018; Tapia-Hernandez, et al., 2019). Insoluble 136 hydrocolloids, such as cellulose or chitin, can also be used to form functional 137 nanoparticles by breaking down bulk materials into smaller fragments using mechanical 138 or chemical methods (Duan, Huang, Lu, & Zhang, 2018; Khalil, et al., 2014). These 139 kinds of organic nanoparticles have been shown to alter the digestibility of 140 macronutrients (such as lipids) using both *in vivo* and *in vitro* studies, which has been 141 linked to various physicochemical and physiological mechanisms (DeLoid, et al., 2018; 142 Liu, Kerr, & Kong, 2019).

143 **3.2. Binding Properties**

144

Food hydrocolloids may contain a number of different functional groups on their

145 surfaces, including non-polar, polar, anionic, and cationic groups, with the number, type 146 and distribution depending on the molecule involved (Phillips, et al., 2009). As a result, 147 they are able to bind to other molecules in their environment through a variety of 148 molecular interactions (Foegeding & Davis, 2011; Stephen, et al., 2006). For instance, 149 hydrocolloids with accessible non-polar patches on their surfaces can bind non-polar 150 molecules through hydrophobic interactions, whereas those with negatively or positively 151 charged patches can bind cationic or anionic molecules through electrostatic interactions, respectively (Blackwood, et al., 2000). As specific examples, acetylated lupin fibers have 152 153 been shown to bind bile salts through hydrophobic interactions (Cornfine, Hasenkopf, 154 Eisner, & Schweiggert, 2010), whereas cationic chitosan has been shown to bind anionic 155 bile salts through electrostatic interactions (Thongngam & McClements, 2005). 156 In general, the binding interactions between two substances ("receptor" and 157 "ligand") can be characterized by a number of parameters: the number of binding sites, the affinity of each binding site, and the molecular origin of the binding interaction (e.g., 158 159 hydrogen bonding, electrostatic forces, hydrophobic forces, and/or van der Waals 160 interactions). In the case of hydrocolloids with multiple binding sites, the sites may act 161 independently or dependently, may be similar or dissimilar from each other, and may be 162 specific or non-specific. The nature of the binding interactions involved depends on the 163 type and concentration of the receptor and ligand, as well as the prevailing environmental 164 conditions (such as pH, ionic strength, and temperature). Consider a simple binding 165 interaction between a receptor and ligand given by the equation:

166

167

$$R + L \leftrightarrow RL \tag{1}$$

168

Thermodynamic analysis of this interaction leads to the following expression relating the
number of bound ligands to the ligand concentration and binding constant (Hulme &
Trevethick, 2010):

172

173
$$[RL] = \frac{[L][R_T]}{[L] + K_d}$$
(2)

174

175 Here, [RL] is the concentration of bound ligand, [L] is the concentration of free ligand,

176 $[R_T]$ is the total receptor concentration, and K_d is the equilibrium dissociation

177 constant, which provides a measure of the tendency of the receptor–ligand complex to 178 dissociate. The fraction of the binding sites (F_B) on the receptor that are bound can then 179 be obtained by rearranging this equation:

180

181
$$F_B = \frac{[L]}{[L] + K_d}$$
 (3)

182

This equation allows one to plot the fraction of ligand molecules bound as a function of the free ligand concentration, provided the dissociation constant of the binding site is known. The fraction of binding sites occupied increases with increasing ligand concentration and decreasing dissociation constant (*i.e.*, increasing binding affinity)

187 (**Figure 1**).

188 In practice, binding interactions involving food hydrocolloids and gastrointestinal 189 components are much more complex than this simple receptor-ligand mechanism, and so 190 more complex mathematical models are required. For instance, the hydrocolloid may 191 have multiple binding sites, the conformation of the hydrocolloid may change after 192 binding, or ligands may bind as molecular complexes (like micelles) rather than 193 individual molecules (monomers). Binding interactions involving hydrocolloids can alter 194 the way that foods behave inside the human gut, which may be either detrimental or 195 beneficial to human nutrition as discussed in Section 5.3.2.

196 **3.3. Surface Activity and Interfacial Properties**

197 A number of important food hydrocolloids are amphiphilic, *i.e.*, they have both non-198 polar and polar segments on the same molecule, *e.g.*, most proteins, gum arabic, and 199 OSA-modified starch. As a result, they can adsorb to oil-water interfaces and form 200 protective coatings (Dickinson, 2003; Garti & Leser, 2001). These types of hydrocolloids 201 are often used to form and stabilize oil-in-water emulsions, which are commonly used as 202 delivery systems for nutrients (McClements, 2015). Many proteins are surface active 203 molecules that can protect oil droplets from aggregating, including those isolated from 204 animal (egg, milk, and meat) and plant (soy, pea, fava bean, and rice) sources. Most 205 polysaccharides are not very surface activity because they have too many hydrophilic

groups on them, but a few of them exhibit strong surface activity because they have some hydrophobic groups attached to the hydrophilic backbone, such as gum arabic, OSAmodified starch, and beet pectin. The nature of the hydrocolloids used to coat the droplets in oil-in-water emulsions can have a pronounced impact on their gastrointestinal fate, by altering the aggregation state of the oil droplets or by altering their surface characteristics (see Section 5.).

The adsorption of a substance to an interface is often described by the *Langmuir adsorption isotherm* (McClements, Bai, & Chung, 2017):

214

215
$$\theta = \frac{\Gamma}{\Gamma_{\infty}} = \frac{c/c_{1/2}}{1 + c/c_{1/2}}$$
(4)

216

Here, θ is the fraction of adsorption sites actually occupied by the substance, Γ is the amount of the substance adsorbed to the surface, Γ_{∞} is the total amount of the substance that could be adsorbed to the surface when it is fully saturated, *c* is the concentration of the substance in the surrounding solution, and $c_{\frac{1}{2}}$ is the concentration of the substance in the surrounding solution when $\theta = \frac{1}{2}$. The surface activity of a substance can be defined as: $SA = \frac{1}{c_{\frac{1}{2}}}$, which provides a quantitative measure of the surface binding affinity. The surface activity of a substance can be related to the free energy of adsorption:

224

225
$$K = \frac{1}{c_{1/2}} = \exp\left(-\frac{\Delta G_{ads}}{RT}\right)$$
(5)

226

227 Here, ΔG_{ads} is the free energy change that occurs when a substance moves from the 228 surrounding fluid to the surface. Thus, the greater (more negative) is the value of ΔG_{ads} , 229 the stronger is the affinity of the substance for the surface. The adsorption free energy is 230 made up of enthalpy and entropy contributions. The enthalpy contributions are due to 231 changes in the magnitude of the molecular interactions as a result of adsorption, such as 232 van der Waals, electrostatic, hydrogen bonding, and hydrophobic and steric interactions. 233 The entropy contributions are due to changes in the number of ways the substance can be 234 arranged in the non-adsorbed and adsorbed states, such as the ability to freely change

235 location, rotate, or undergo conformational changes. In some cases, the various 236 contributions to ΔG_{ads} can be calculated using mathematical models (Norde, 2011).

237 The adsorption of hydrocolloids to the interfaces in multiphase foods (such as 238 emulsions or starch suspensions) can alter their gastrointestinal fate. These effects 239 depend on the nature of the interfacial layers formed, such as their composition, structure, 240 thickness, charge, and polarity (McClements, et al., 2017). For instance, the interfacial 241 layer properties may impact the aggregation state of fat droplets within the GIT, which 242 would alter the surface area available for digestive enzymes to adsorb to (Zeeb, Lopez-243 Pena, Weiss, & McClements, 2015). Alternatively, they may form a steric barrier that 244 inhibits the ability of the digestive enzymes to access the macronutrients (McClements & 245 Li, 2010). The potential impact of interfacial layers on the GIT of foods is discussed in a 246 later section (Section 5.3.3).

247 **3.4. Thickening Properties**

248 Some hydrocolloids are highly effective at increasing the shear viscosity of aqueous 249 solutions due to their large molecular dimensions (Bai, et al., 2017). The viscosity of a 250 polymer solution is determined by the fluid flow around the polymer molecules, since 251 this generates additional friction within the fluid (energy dissipation). Thus, 252 hydrocolloids that have highly extended structures tend to be more effective at thickening 253 aqueous solutions because of their ability to perturb the fluid flow more strongly (Figure 254 2). The ability of a hydrocolloid to increase the viscosity of an aqueous solution can be 255 related to its volume ratio (R_V) , which is the effective volume occupied by the molecule 256 in solution (polymer chain plus entrained water) divided by the volume occupied by only 257 the polymer chain alone (Bai, et al., 2017). The greater the volume ratio, the more 258 effective is the hydrocolloid at increasing the viscosity. The volume ratio tends to 259 increase with increasing molecular weight, decreasing branching, and increasing 260 extension. Thus, rod-like molecules (like xanthan) have higher viscosities than random 261 coil molecules (like hot gelatin), which in turn have higher viscosities than compact 262 globular proteins (like whey or egg proteins).

A relatively simple semi-empirical equation can be used to predict the ability of food hydrocolloids to thicken aqueous solutions (Bai, et al., 2017):

265

266
$$\eta = \eta_1 \left(1 - \frac{\phi_E}{\phi_C} \right)^{-2} \tag{6}$$

267

In this equation, η is the shear viscosity of the hydrocolloid solution, η_1 is the shear viscosity of the aqueous solution surrounding the hydrocolloids, ϕ_E is the *effective volume fraction* of the hydrocolloid molecules in solution, and ϕ_C is a critical packing fraction (\approx 0.57). This latter term corresponds to the volume fraction of spheres that can be packed into a suspension before they become tightly packed, after which there is a steep rise in the viscosity of the suspension. To a first approximation, the effective volume fraction occupied by hydrocolloid molecules in solution is given by (Bai, et al., 2017):

276
$$\phi_E = \frac{4}{3}\pi r_H^3 \left(\frac{cN_A}{M}\right) \tag{7}$$

277

In this expression, $r_{\rm H}$ is the hydrodynamic radius of the hydrocolloids, c is the hydrocolloid concentration, $N_{\rm A}$ is Avogadro's number, and M is the hydrocolloid molecular weight. As shown in **Figure 3**, these equations predict that the ability of a hydrocolloid to thicken a solution rises as the concentration and effective volume of the hydrocolloid molecules increases.

A simple expression for the critical viscosity concentration (CVC), which is the hydrocolloid concentration where there is a steep rise in viscosity due to overlap of the polymer chains, has been derived from the above equations, which relates the thickening power to the radius of hydration of a hydrocolloid (Grundy, McClements, Ballance, & Wilde, 2018):

288

$$289 \qquad CVC = 21 \times \frac{M}{r_H^3} \tag{8}$$

290

Here, CVC is given in wt% when the molecular weight of the hydrocolloid is given in kg mol⁻¹ and the radius of hydration is given in nanometers. The thickening power can be conveniently defined as the reciprocal of CVC. Hence, the thickening power increases as the radius of hydration increases (for the same molecular weight), *i.e.*, the hydrocolloid molecules become more extended. Hence, long rigid hydrocolloids (like xanthan gum)
are much more effective thickening agents than compact globular hydrocolloids (like
whey protein) (Figure 3).

Overall, these equations are useful for understanding the potential for different hydrocolloids to thicken aqueous solutions. The ability of hydrocolloids to thicken aqueous solutions can be utilized to control the gastrointestinal fate of foods. For instance, these hydrocolloids can be used to increase the viscosity of the gastrointestinal fluids in the stomach or small intestine, thereby slowing down mixing and mass transfer processes and altering the release of flavors or nutrients (see Section 5.3.1).

304 3.5. Gelling Properties

305 Many food hydrocolloids are capable of forming hydrogels consisting of a network 306 of cross-linked biopolymer molecules that entrap water through capillary forces (Phillips, 307 et al., 2009). The nature of the gels formed, such as their rheology, appearance, and 308 response to environmental changes, depend on the type of biopolymers and cross-links 309 employed. Thus, it is possible to form hydrogels with different optical properties (clear, 310 turbid, opaque), rheological properties (soft to hard, rubbery to brittle), setting properties 311 (cold-, hot-, salt-, or enzyme-set), and digestibility (indigestible or digestible). Hydrogels 312 can be formed within a food product prior to consumption or they can be formed within 313 the human body after ingestion of the hydrocolloids (Norton & Frith, 2001). The 314 dimensions, pore size, and degradability of hydrogels can be tuned so as to control the 315 retention, stability, and release of nutrients within the gastrointestinal tract (McClements, 316 2017). Hydrocolloids can be used to form macroscopic or microscopic gels depending on 317 the application. A number of examples of using hydrocolloids for this purpose is given 318 later.

The elastic modulus of a polymer gel depends on the nature, concentration, and interactions of the polymer molecules in the gel network (Vilgis, 2015). A variety of mathematical models have been developed to relate the structural properties of polymer gels to their rheological properties (Gabriele, de Cindio, & D'Antona, 2001; Rubinstein, Colby, Dobrynin, & Joanny, 1996). These models predict that the elastic modulus depends on polymer concentration, crosslinking density, and bond strength. In addition, theoretical models have been developed to describe the elastic modulus of polymer gels

326 containing embedded particles (Fu, Feng, Lauke, & Mai, 2008). These theories show that 327 the size, concentration, and interactions of the colloidal particles with the polymer 328 network impact their rheological properties, such as stiffness, strength and toughness. 329 These models can sometimes be useful for predicting the behavior of food gels before 330 consumption, but they are usually too simple to understand the complex behavior of food 331 gels within the mouth during mastication and in the stomach and small intestine when 332 they breakdown through mechanical, chemical, or enzymatic mechanisms. Instead, it is 333 typically more important to empirically establish the influence of GIT factors such as pH, 334 ionic strength, bile salt concentration, and enzyme activity on the properties of a 335 hydrocolloid gel, e.g., swelling, shrinking, fragmentation, disassembly, or erosion 336 (Figure 4). These processes can be controlled to design colloidal delivery systems that 337 can trigger or control the release rate of bioactive agents in different regions of the GIT 338 (Section 6). As an example, researchers have created "soft" and "hard" whey protein gels 339 by varying the nature of the gelation mechanism used to produce them (Guo, et al., 340 2015). The authors found that under simulated gastric conditions (with pepsin) the soft 341 gels disintegrated more rapidly, and led to faster gastric emptying, which could have 342 important nutritional consequences. In other studies, it has been shown that the size of the 343 pores in food gels impacts the rate of digestion of encapsulated macronutrients. For 344 instance, it has been shown that the rate of lipid digestion is reduced when lipid droplets 345 are encapsulated within hydrogel matrices, either made from proteins or polysaccharides, 346 because the lipase has to penetrate through the gel network before accessing the lipid 347 surfaces (Li, Hu, Du, Xiao, & McClements, 2011a; Sarkar, et al., 2015). In these 348 examples, the rate of lipid digestion decreases as the mesh size of the polymer network 349 decreases because it is more difficult for the digestive enzymes to penetrate.

350

3.6. Alteration of Mass Transport

It is often assumed that hydrocolloids that thicken or gel aqueous solutions will greatly slow down mass transport processes, but this depends on the nature of the molecules involved. It is important to distinguish between the *macro*-viscosity and the *micro*-viscosity of a hydrocolloid solution (Desmidt & Crommelin, 1991; Gulnov, Nemtseva, & Kratasyuk, 2016). The macro-viscosity is usually measured on bulk samples using conventional rheological instruments, such as shear viscometers or

357 dynamic shear rheometers. It depends on the resistance to flow of a material when a 358 macroscopic force is applied. Conversely, the micro-viscosity is related to the resistance 359 to movement that small molecules or small particles experience on a nano- or micro-360 scale, which is usually reflected by measuring their diffusion coefficient. The micro-361 viscosity can be measured using various methods, including particle tracking, dynamic 362 light scattering, and fluorescence spectroscopy (Furst, Squires, Furst, & Squires, 2017; 363 Vysniauskas & Kuimova, 2018). The micro- and macro-viscosity are fairly similar for 364 simple solutions containing small molecules, such as water or a sugar solution. However, 365 they can be orders of magnitude different for more complex solutions containing 366 hydrocolloids, such as starch or gelatin (Gulnov, et al., 2016). A small molecule or 367 particle may be able to travel unhindered through the spaces between a large hydrocolloid 368 molecule and so experiences a local viscosity close to that of the pure solvent (usually 369 water), even though the macro-viscosity can be very high (Basaran, Coupland, & 370 McClements, 1999). On the other hand, the movement of large molecules or particles 371 may be hindered when their dimensions are similar to or larger than the pore size in the 372 hydrocolloid network (Burla, Sentjabrskaja, Pletikapic, van Beugen, & Koenderink, 373 2020). This phenomenon is important when considering the diffusion of bile salts or 374 enzymes through gastrointestinal fluids that may contain hydrocolloids that thicken or gel 375 them, or when considering the release of nutrients from gelled phases or microgels 376 (McClements, 2017).

To a first approximation, the movement of non-interacting small particles (molecules, ions, or nanoparticles) through the pores in a spherical microgel can be described using the expression below (Crank, 1975; McClements, 2017):

380

381

$$\Phi = \frac{M(t)}{M(\infty)} = 1 - exp\left[-\frac{1.2D\pi^2}{Kr^2}t\right]$$
(9)

382

Here, Φ is the fraction of the particles that have diffused into or out of the microgels in time *t*, M(t) and $M(\infty)$ are the particle concentrations in the microgels at time *t* and at equilibrium (infinite time), *r* is the radius of the microgels, *D* is the diffusion coefficient of the particles through the polymer network inside the microgels, and *K* is the equilibrium partition coefficient of the particles between the microgels and surrounding fluid. The following expression has been derived to predict the hindered diffusion of
particles through polymer networks (Chan & Neufeld, 2009; Zhang & Amsden, 2006):

391
$$D_{gel} = D_w \exp\left(-\pi \left(\frac{r_H + r_f}{\xi + 2r_f}\right)\right)$$
(10)

392

Here, D_{gel} and D_w are the diffusion coefficients of the particles through the polymer network and through pure water, respectively, r_H is the hydrodynamic radius of the particles, r_f is the cross-sectional radius of the polymer chains in the polymer network, and ζ is the diameter of the pores in the polymer network. The diffusion coefficient of the particles through the water can be related to their dimensions using the following expression (assuming they are roughly spherical):

399

$$D_{\rm w} = k_{\rm B} T / 6\pi \eta r_{\rm H} \tag{11}$$

401

402 Here, $k_{\rm B}$ is Boltzmann's constant, T is absolute temperature, and η is the viscosity of the 403 surrounding liquid (usually water). These equations can be used to better understand the 404 factors that impact the release of components from hydrocolloid-based viscous solutions 405 or gels present in the gastrointestinal tract through diffusion, such as the release of flavors 406 in the mouth, the penetration of enzymes into food matrices in the stomach, or the release 407 of bioactive agents from microgels in the small intestine. These equations predict that the 408 mass transport rate of a flavor, enzyme, or bioactive agent should increase as the pore 409 size in the polymer network increases. This knowledge can then be used to design 410 hydrogel-based foods that slow down the digestion of macronutrients or that control the 411 release of flavors or nutrients in the body.

Finally, it should be noted that the macro-viscosity may also impact the mass transport of foods and gastrointestinal components by altering mixing processes, which could alter nutrient digestion (Farres, Moakes, & Norton, 2014). For instance, if there are large clumps of gelled material in the GIT that contain macronutrients, the digestive enzymes may have to diffuse through these clumps to get to the lipids, proteins, or starches inside, thereby retarding their hydrolysis.

418 **3.7. Water-holding Capacity and Gastrointestinal Transport**

419 Many food hydrocolloids have good water-holding capacity (WHC) due to their 420 ability to hold water through a capillary mechanism (Blackwood, et al., 2000; Stevenson, 421 Dykstra, & Lanier, 2013). As a result, they can alter the rheology of the partially digested 422 foods passing through the GIT. In particular, the presence of dietary fibers can increase 423 the amount of water trapped in the stool as it passes along the colon, thereby making it 424 softer and easier to move, which helps to prevent constipation and improve gut health. 425 To a first approximation, the Laplace pressure of a porous material is given by the 426 following equation (Stevenson, et al., 2013):

427

428

 $\Delta P = 2\gamma \cos \theta/r$

429

430 Here, ΔP is the capillary pressure, γ is the water-air surface tension, θ is the contact angle 431 at the water-polymer interface, and *r* is the radius of the pores. The water holding 432 capacity (WHC) of a porous food material is related to its ability to hold water against 433 some external force, such as gravity or applied pressure. The WHC typically increases as 434 the number of pores increases and the size of the pores decreases.

(12)

(15)

Thermodynamic models have been developed to relate the water holding capacity of polymer-based foods (such as meats or vegetables) to their chemical and structural properties (van der Sman, 2013; van der Sman, Paudel, Voda, & Khalloufi, 2013). These models relate the WHC of polymeric food materials to three main factors: (i) polymerwater mixing; (ii) impact of ion – polymer interactions; and (iii) elastic contributions due to deformation of the gel matrix. The theory is developed in terms of the swelling pressure:

442

$$443 \qquad \Pi_{\rm S} = \Pi_{\rm M} + \Pi_{\rm I} + \Pi_{\rm E} \tag{13}$$

445
$$\Pi_M = +\frac{kT}{v_W} [\ln(1-\phi) + \phi + \chi \phi^2]$$
(14)

$$\Pi_I = +\frac{RT}{v_W} [a_{w,I}]$$

447
$$\Pi_E = -\frac{RT}{v_W} N_C \phi_0 \left[\frac{1}{2} \left(\frac{\phi}{\phi_0} \right) - \left(\frac{\phi}{\phi_0} \right)^{1/3} \right]$$
(16)

448

Here, *R* is the gas constant, *T* is the absolute temperature, v_W is the molar volume of the water, ϕ is the volume fraction of the polymer molecules, χ is the Flory–Huggins interaction parameter, $a_{w,I}$ is the water activity of the added ions, *N*_c depends on the number density of polymer crosslinks, and ϕ_0 is the volume fraction of polymer at crosslinking (van der Sman, et al., 2013).

454 Mathematical models have also been developed to relate the microstructure and 455 surface properties of colloidal gels to their water holding characteristics (Chang & 456 Cheng, 2018; Smagin, 2018; Smagin, et al., 2019). These models were originally 457 developed to predict the retention of water by colloidal soils but should also be suitable 458 for application to colloidal foods. As expected, they predict that the water holding 459 capacity of a material increases as the pore size decreases and the surface area increases. 460 This relationship between capillary forces and WHC has been demonstrated empirically 461 for gels made from non-food and food polymers: water increases as pore size decreases 462 (Stevenson, et al., 2013). A better understanding of the physicochemical basis for the 463 WHC of the gastrointestinal contents may lead to the design of functional foods that can 464 prevent constipation and improve gut function.

465 **3.8. Digestibility and Fermentability**

466 Differences in hydrocolloid digestibility in different regions of the gastrointestinal 467 tract can be employed to create smart colloidal delivery systems capable of releasing 468 nutrients to targeted regions within the human gut (Figure 5). Some food hydrocolloids 469 are digestible within the upper gastrointestinal tract, such as many proteins and starches 470 (McClements, 2017). For instance, starch-based particles can be digested within the 471 mouth by amylase and release their payload there. Conversely, protein- or lipid-based 472 ones are digested in the stomach and small intestine and are therefore more suitable for 473 delivery of payloads to these regions. Other food hydrocolloids (dietary fibers) are 474 indigestible in the mouth, stomach, and small intestine, but are fermented by gut bacteria 475 once they reach the colon. These fermentable hydrocolloids can be used to assemble 476 dietary fiber-based particles that can deliver nutrients to the colon (McClements, 2017),

477 or they can serve as prebiotics to promote the growth of beneficial bacteria in the gut478 (Roberfroid, et al., 2010; Wang, 2009).

The digestibility and fermentability of food hydrocolloids are therefore important
characteristics that can be used to control the gastrointestinal fate of nutrients, thereby
improving human health and wellbeing. Potential applications of hydrocolloids for this
purpose are discussed later.

483 **4.** The physiology and physiochemistry of the human gut

484 Before examining the impact of food hydrocolloids on the behavior of foods within 485 the gastrointestinal tract it is useful to briefly discuss the digestion and absorption of 486 nutrients and the importance of controlling these processes. Traditionally, food scientists 487 have been primarily concerned with creating shelf-stable foods with desirable 488 physicochemical and sensory properties. More recently, there has been interest in also 489 designing foods so as to control their behavior within the human gut, so as to produce 490 desirable nutritional or physiological effects, such as increased satiety, enhanced 491 bioavailability, modulated pharmacokinetics, or targeted release (Chung, Smith, Degner, 492 & McClements, 2016; Krop, et al., 2018; Sarkar, Ye, & Singh, 2017a). The design of 493 these kinds of functional foods requires knowledge of the different regions of the 494 gastrointestinal tract, the multiscale dynamic processes occurring there, and how they 495 alter food properties (Bornhorst, Gouseti, Wickham, & Bakalis, 2016). Moreover, food 496 and nutrition researchers are utilizing concepts and techniques developed in the 497 pharmaceutical industry for drug delivery to better understand and control the 498 gastrointestinal fate of foods (Gleeson, Ryan, & Brayden, 2016; Nowak, Livney, Niu, & 499 Singh, 2019; Sarkar & Mackie, 2020). In particular, standardized in vitro and in vivo 500 gastrointestinal and pharmacokinetic models are being increasingly adopted by food 501 researchers. In this section, a brief overview of the journey of foods through the human 502 gut is therefore given.

503 **4.1. Mouth**

504 Foods enter the human body through the mouth, whose initial purpose is to sample 505 foods to determine whether they should be further processed or spat out. If the food is 506 deemed acceptable, then the mouth converts it into a form that is suitable for swallowing

507 (Singh, Ye, & Horne, 2009). For fluid foods, this simply involves holding the food

- 508 within the mouth before swallowing but for solid foods this may also involve a
- 509 considerable amount of chewing to breakdown the structure and mix the food with the
- 510 saliva. Saliva is a complex biological fluid with a pH around neutral that contains
- 511 minerals, enzymes (amylase), antimicrobials, and mucin, which is a natural hydrocolloid
- that lubricates the oral surfaces and helps foods slip down the esophagus in the form of a
- 513 bolus (Dawes, et al., 2015; Sarkar, Xu, & Lee, 2019). The time foods spend in the mouth
- 514 varies from a few seconds to a few minutes depending on their textural characteristics,
- 515 *e.g.*, low viscosity fluids *versus* hard chewy solids. It should be noted that hydrocolloids
- 516 may make foods feel creamy, watery, slimy, or gritty within the mouth depending on their
- 517 molecular and physicochemical characteristics. Consequently, the impact of
- 518 hydrocolloids on mouthfeel is an important consideration when developing functional
- 519 foods (Shewan, Pradal, & Stokes, 2020; Upadhyay, Aktar, & Chen, 2020).

520 **4.2. Stomach**

521 The stomach is a muscular cavity that contains highly acidic (pH 1-3) and 522 enzymatically-active gastric fluids (Guo, Ye, Singh, & Rousseau, 2020; Singh, et al., 523 2009). The low pH of the stomach helps protect the body from harmful microbes that 524 might be present in the ingested food. In addition, the stomach is designed to breakdown 525 any remaining food structures through a combination of mechanical (churning), chemical 526 (hydrochloric acid), and enzymatic (gastric lipase and pepsin) processes. On average, 527 solid foods remain in the stomach for about 2 hours, but there is a broad range of 528 residence times in practice. Before foods can enter the small intestine, they must pass 529 through the pylorus sphincter, which is about 2 mm in diameter.

530 **4.3. Small intestine**

After passing through the pylorus sphincter, the partially digested food from the stomach ("chyme") enters the initial stages of the small intestine (duodenum). Alkaline pancreatic juices containing bile salts and digestive enzymes are squirted into the chyme causing its pH to become closer to neutral and initiating further hydrolysis of the macronutrients (Bohn, et al., 2018; Norton, Espinosa, Watson, Spyropoulos, & Norton,

536 2015; Singh, et al., 2009). Here, starches are broken down to glucose by amylases,

537 proteins are broken down to peptides and amino acids by proteases, and lipids are broken 538 down to monoglycerides and free fatty acids by lipases (Boland, Golding, & Singh, 539 2014). The water-soluble digestion products diffuse through the intestinal fluids to the 540 surfaces of the epithelium cells where they can be absorbed by active transporters in the 541 cell membranes or by passive diffusion through the T-junctions that hold neighboring 542 cells together. Small water-insoluble digestion products, such as fatty acids and 543 monoglycerides, may be incorporated into mixed micelles that then carry them to the 544 epithelium cells for absorption, where they are then re-assembled and packaged into 545 lipoproteins that carry them through the bloodstream to other tissues (Ko, Qu, Black, & 546 Tso, 2020). Large insoluble matter, such as dietary fibers, calcium soaps, and non-547 digestible lipids may not be absorbed within the small intestine, and so they move further 548 down the GIT to the colon. The rate and degree of macronutrient digestion determines 549 the pharmacokinetic (PK) profile of the nutrient metabolites in the bloodstream, such as 550 glucose, lipoproteins, and peptides (Figure 7). The PK profile influences the total number 551 of calories obtained from a food, as well as hormonal and metabolic responses that affect 552 satiety, appetite, and insulin sensitivity.

553 **4.4. Colon**

554 Some food hydrocolloids are not digested or only partially digested within the upper 555 gastrointestinal tract because they are resistant to hydrolysis by the enzymes secreted 556 there (Grabitske & Slavin, 2009; Kumar, Sinha, Makkar, de Boeck, & Becker, 2012; 557 Lattimer & Haub, 2010). This might occur because they contain covalent linkages (such 558 as certain types of glycosidic bonds) that cannot be hydrolyzed by the digestive enzymes 559 or because they are physically protected from digestion (e.g., because they are trapped560 inside indigestible matrices or surrounded by indigestible coatings). Hydrocolloids that 561 are not digested in the upper GIT may be hydrolyzed by enzymes secreted by the 562 microbes residing in the colon. One of the main fermentation products arising from the 563 digestion of dietary fibers are short chain fatty acids (SCFAs), such as acetate, propionate 564 and butyrate, which act as a fuel source for the colonic enterocytes, as well as signaling 565 molecules with the host. As a result, they play an important role in maintaining gut 566 health. The type and amount of SCFAs and other metabolites produced in the colon 567 impacts the nature and diversity of the gut bacteria, which plays a major role in human

health and wellbeing (Carlson & Slavin, 2016; Tabernero & de Cedron, 2017).

569 Undigested proteins reaching the colon may also serve as a source of nutrients for the

570 bacteria living in the colon. Controlling the types of hydrocolloids reaching the colon

571 through food design approaches may therefore be important for controlling human health

572 by modulating the gut microbiome.

573 5. Impact of Hydrocolloids on Nutrient Gastrointestinal Fate

574 **5.1. Modulation of Mouthfeel and Flavor**

575 Many hydrocolloids play an important role in the mouthfeel of both natural and processed foods, which is mainly due to their ability to alter the rheological properties of 576 577 the food and saliva (Mosca & Chen, 2017; Wang & Chen, 2017), as well as coating and 578 lubricating the surfaces of the mouth (Sarkar, Andablo-Reyes, Bryant, Dowson, & 579 Neville, 2019; Sarkar, et al., 2017a). The nature of these effects depends on the molecular 580 characteristics of the hydrocolloids, such as their molecular weight, conformation, 581 polarity, and charge, because this impacts their ability to alter the fluid flow and to 582 interact with saliva components and oral surfaces (Stokes, Boehm, & Baier, 2013). Many 583 hydrocolloids greatly increase the viscosity of the bolus, which can either lead to an 584 unpleasant gumminess/slipperiness or a desirable thickness/creaminess, depending on the 585 nature of the effect (Upadhyay, et al., 2020; van Vliet, van Aken, de Jongh, & Hamer, 586 2009). Some proteins have been reported to have an astringent mouthfeel, which can be 587 attributed to their tendency to bind to mucin and form insoluble complexes (Celebioglu, 588 Lee, & Chronakis, 2020). As a result, the mucin is no longer able to lubricate the oral 589 surfaces effectively (Fabian, Beck, Fejerdy, Hermann, & Fabian, 2015). Some peptides, 590 formed by hydrolyzing proteins, have a bitter taste, which can reduce the desirable flavor 591 profile of foods (Fu, Chen, Bak, & Lametsch, 2019). Consequently, it is important to 592 prevent these peptides from being present in the mouth or to design food matrices that 593 mask their bitterness.

The presence of hydrocolloids within the mouth may also alter the flavor profile of
foods (Hollowood, Linforth, & Taylor, 2002; Taylor, 1996; Taylor & Linforth, 1996).
Hydrocolloids alter the mixing of foods with saliva, as well as the movement of taste and
aroma molecules from the food to flavor receptors in the mouth and nose. The ability of

598 hydrocolloids to modulate flavor release depends on numerous factors, with the most 599 important being their ability to bind certain kinds of flavor molecules and to increase the 600 viscosity of the oral contents. Many proteins and starches have accessible non-polar 601 domains on their surfaces that can bind non-polar aroma molecules, which decreases the 602 amount reaching the nasal cavity, thereby reducing flavor intensity (Kuhn, Considine, & 603 Singh, 2006; Viry, Boom, Avison, Pascu, & Bodnar, 2018). Hydrocolloids may also 604 delay the release of both taste and aroma molecules due to their ability to hinder their 605 movement from the ingested food to the saliva and into the gas phase above the food 606 (Hollowood, et al., 2002). Consequently, the type, concentration, and interactions of 607 hydrocolloids in foods impacts flavor perception, which can be utilized to create foods 608 with desirable mouthfeel and flavor release profiles.

609 **5.2. Modulation of Gastrointestinal Transit**

610 5.2.1. Gastric motility and emptying

611 The length of time a food remains in the stomach and the speed at which it is broken 612 down due to mechanical, enzymatic, and chemical processes affects the rate of nutrient 613 release and absorption (Ratanpaul, Williams, Black, & Gidley, 2019). In turn, this 614 influences the timing of hormone release in response to food ingestion, which impacts 615 hunger, satiety, and satiation, and therefore the frequency and quantity of food consumed 616 (Bruen, O'Halloran, Cashman, & Giblin, 2012; Halford & Harrold, 2012). Some 617 hydrocolloids are able to increase the viscosity of the gastric fluids, which slows gastric 618 emptying and macronutrient digestion (Guo, et al., 2020; Qi, Al-Ghazzewi, & Tester, 619 2018). This delay in gastric emptying may increase the feelings of satiety and satiation, 620 thereby reducing the total amount of calories consumed, leading to health benefits by 621 reducing body weight. Moreover, they may be able to control hormone levels thereby 622 improving metabolic health. The impact of food hydrocolloids on gastric motility and 623 emptying may therefore influence chronic diseases related to overeating, such as obesity, 624 diabetes, and heart disease. Slower gastric emptying may also help to prevent spikes in 625 the concentration of sugars or lipids in the bloodstream, which can enhance metabolic 626 health.

627 *5.2.2. Colonic flow*

628 The speed that non-adsorbed foods move through the colon may also have important 629 health effects. As mentioned earlier, the main factor impacting the rheology of the 630 gastrointestinal contents in the colon is the absorption of water, which depends on 631 capillary forces generated by the hydrocolloid network inside (Stevenson, et al., 2013). 632 Dietary fibers form a 3D-network within the gastrointestinal contents in the colon, which 633 can entrain large quantities of water through capillary forces. As a result, the rheological 634 properties of the gastrointestinal contents in the colon are highly dependent on the type 635 and amount of dietary fiber present, which influences the speed and ease at which they 636 can pass through the lower GIT (Blackwood, et al., 2000). In principle, a diet low in 637 fiber leads to a more rigid solid-like mass that moves slowly, whereas a diet high in fiber 638 leads to a more fluid-like mass that passes through more rapidly, thereby reducing 639 constipation. Nevertheless, not all dietary fibers are effective at reducing constipation, 640 with many fibers having little or no effect (Gelinas, 2013). Indeed, in a recent review it 641 was stated that "to exert a laxative effect, fiber must: (1) resist fermentation to remain 642 intact throughout the large bowel and present in stool, and (2) significantly increase stool 643 water content and stool output, resulting in soft/bulky/easy-to-pass stools." (McRorie & 644 Chey, 2016). As a specific example, a study with preterm infants showed that 645 supplementation of their meals with prebiotic oligosaccharides reduced stool viscosity 646 and reduced transit times (Mihatsch, Hoegel, & Pohlandt, 2006). In future, more research 647 is required to identify those hydrocolloids that are most effective at increasing the rate of 648 transit of foods through the colon.

649

650 **5.3. Modulation of Macronutrient Digestion and Absorption**

The presence of hydrocolloids in foods can modulate the digestion and/or absorption of macronutrients, such as fats, starches, and proteins. In some cases, they decrease the rate and extent of these processes, whereas in other cases they may have little effect, or even the opposite effect, depending on their nature and concentration. In this section, some of the major mechanisms that hydrocolloids can modulate nutrient digestion and absorption are highlighted.

657 5.3.1. Rheological modification of gastrointestinal fluids

658 The presence of hydrocolloids within a food may alter the rheological properties of 659 the gastrointestinal fluids, which then impacts the digestion and absorption of 660 macronutrients. For instance, some hydrogels can form highly viscous solutions or gels 661 within the gastrointestinal tract (Farres, et al., 2014). A study with an in vitro GIT model, 662 which included segmentation forces in the small intestine, showed that adding a 663 thickening agent (guar gum) to the simulated intestinal fluids reduced the rate of glucose 664 release after starch hydrolysis (Tharakan, et al., 2010). Adding this kind of hydrocolloid 665 may therefore be useful for designing foods for diabetics, who would benefit from a low 666 and sustained level of glucose within their bloodstream, rather than experiencing large 667 spikes. Hydrocolloids may also thicken or gel the gastric fluids, which reduces the 668 mixing of the different constituents within the stomach (Farres, et al., 2014). As a result, 669 it is more difficult for the digestive enzymes to reach the macronutrients and hydrolyze 670 them. For instance, fat droplets or starch granules may be trapped within highly viscous 671 or gelled regions within the stomach that enzymes can only reach slowly by diffusing 672 through the biopolymer network.

673 5.3.2. Binding interactions

674 As mentioned earlier, the binding of gastrointestinal constituents to ingested food 675 hydrocolloids may alter the way foods behave inside the human gut. Understanding and 676 controlling these interactions may therefore be used to improve human nutrition. In 677 general, binding may occur due to a number of different kinds of molecular forces, 678 depending on the nature of the molecules involved and the gastrointestinal environment 679 (such as pH and ionic composition). For instance, binding may occur due to electrostatic, 680 hydrophobic, hydrogen bonding, or van der Waals interactions, or some combination of 681 these forces (Israelachvili, 2011). Consequently, it is important to elucidate the primary 682 types of molecular interactions that each kind of hydrocolloid can participate in, and to 683 identify the key molecular features that lead to these interactions, such as the number and 684 location of charged, polar, or non-polar groups.

685 *In vitro* studies have shown that nanocrystalline cellulose (NCC) can inhibit the 686 activity of α -amylase and α -glucosidase in model foods containing cooked potato starch 687 and protein (Nsor-atindana, Yu, Goff, Chen, & Zhong, 2020). The degree of inhibition 688 was reported to increase as the dimensions of the NCC decreased, which was probably 689 due to the increase in surface area. The authors postulated that the enzymes bound non-690 specifically to the NCC, which reduced its activity against the starch. Other *in vitro* 691 studies have shown that α -amylase activity is inhibited in the presence of cellulose, 692 which was attributed to the binding of the enzyme to this dietary fiber (Dhital, Gidley, & 693 Warren, 2015). This type of hydrocolloid may therefore be useful for slowing down the 694 digestion of starches in the human GIT, thereby reducing glucose spikes, which may 695 again be useful for designing foods for diabetics.

696 Cationic chitosan can bind to anionic fatty acids or bile acids in the small intestine 697 through electrostatic interactions, which causes them to precipitate, thereby reducing the 698 bioaccessibility of oil-soluble vitamins, like vitamin D (Tan, et al., 2020). Similarly, 699 anionic alginate molecules can bind to cationic calcium anions in the stomach or small 700 intestine, which can alter the gastrointestinal fate of nutrients. For example, in vitro 701 experiments have shown that alginate can retard lipid digestion by binding to calcium 702 ions in the simulated small intestine (Hu, et al., 2010; Qin, Yang, Gao, Yao, & 703 McClements, 2016). The origin of this effect is that free calcium ions are normally 704 needed to precipitate long-chain fatty acids that accumulate at the lipid droplet surfaces 705 during digestion. If there are no free calcium ions available to remove these fatty acids, then lipase may not be able to reach the underlying triglycerides. Human studies have 706 707 shown that ingestion of high alginate levels can increase the fraction of free fatty acids 708 excreted in the stool, which was attributed to the formation of alginate gels that trapped 709 them (Sandberg, et al., 1994). The binding of bile salts to dietary fibers in the small 710 intestine can reduce blood cholesterol levels, since they are excreted in the feces, so the 711 body has to utilize endogenous cholesterol to synthesize more bile salts (Singh, Metrani, 712 Shivanagoudra, Jayaprakasha, & Patil, 2019). In vitro isothermal titration calorimetry 713 (ITC) measurements have confirmed that bile salts can bind to chitosan mainly through 714 electrostatic interactions (Thongngam, et al., 2005). Hydrocolloids that can strongly bind 715 bile salts may therefore be useful for reducing the cholesterol levels in individuals who 716 are prone to heart disease.

717 5.3.3. Aggregation state

718 The presence of hydrocolloids may alter the aggregation state of macronutrient 719 particles (such as fat droplets, starch granules, or protein particles) within the stomach or 720 small intestine, thereby altering the surface area that is exposed to digestive enzymes in 721 the gastrointestinal fluids (McClements, Decker, Park, & Weiss, 2009; Zhang, Zhang, 722 Zhang, Decker, & McClements, 2015). Experiments have shown that the rate of 723 macronutrient digestion increases as their surface area increases (particle size decreases). 724 For instance, the rate of lipid digestion in emulsions has been reported to increase as the 725 surface area of lipids exposed to lipases increases in simulated gastrointestinal studies (Li 726 & McClements, 2010b; Salvia-Trujillo, Qian, Martin-Belloso, & McClements, 2013). 727 Similarly, the digestion rate of starch granules and protein particles by amylases or 728 proteases has been reported to increase as their surface area increases (Tamura, Okazaki, 729 Kumagai, & Ogawa, 2017; Xing, et al., 2016).

730 If macronutrient particles form tightly packed clusters, then it is more difficult for the 731 enzymes to access the surfaces of the particles in the interior of the clusters (Figure 6), 732 which slows down digestion. Hydrocolloids can either promote or inhibit macronutrient 733 digestion by inhibiting or promoting particle aggregation within the GIT. For instance, 734 pectin has been shown to inhibit the flocculation of gelatin-coated oil droplets in the 735 stomach, which increased the subsequent rate of lipid digestion in the small intestine by 736 increasing the surface area of lipid droplets exposed to the lipase (Zeeb, Weiss, & 737 McClements, 2015). Similarly, the presence of xanthan gum or pectin (two anionic 738 polysaccharides) was shown to increase the digestibility of hydrolyzed rice glutelin-739 stabilized fish oil-in-water emulsions, which was attributed to their ability to reduce 740 droplet aggregation in a simulated GIT (Xu, Sun, & McClements, 2020). Conversely, the 741 presence of chitosan was shown to promote the flocculation of lipid droplets in the 742 stomach and small intestine, which decreased the rate of lipid hydrolysis (Qin, et al., 743 2016). Consequently, it may be possible to modulate the digestibility of macronutrients 744 within the human gut by adding hydrocolloids that either promote or inhibit their 745 aggregation. Hydrocolloids that inhibit macronutrient digestion through this mechanism 746 may be useful for creating functional foods that suppress sugar or lipid spikes in the 747 blood. Conversely, hydrocolloids that promote digestion may be used to increase the

748 bioavailability of beneficial macronutrients, such as proteins.

749 5.3.4. Interfacial modification and embedding

750 Macronutrient digestion may also be modulated by the properties of any coatings 751 surrounding the fat, starch, or protein particles within a food (Figure 6). These thin 752 coatings can inhibit the ability of digestive enzymes to access the surfaces of the 753 macronutrients, thereby delaying their digestion (McClements, et al., 2010). For 754 instance, anionic polysaccharides may form coatings around cationic lipid droplets in the 755 stomach, which reduces the ability of lipases to access the emulsified lipids in the small 756 intestine (Araiza-Calahorra & Sarkar, 2019; Qin, et al., 2016). A recent study showed 757 that lipid droplets coated by a 3-layer coating (lactoferrin, alginate and polylysine) had a 758 higher digestibility and carotenoid bioaccessibility than those coated by 1- or 2-layers, 759 which was attributed to the ability of the 3-layer coatings to protect the droplets from 760 aggregation in the simulated GIT (Gasa-Falcon, Acevedo-Fani, Oms-Oliu, Odriozola-761 Serrano, & Martin-Belloso, 2020). Another recent study showed that coating whey 762 protein-coated oil droplets with chitosan increased the bioaccessibility of curcumin in a 763 simulated GIT model, as well as the uptake of curcumin by a cell culture model (Gasa-764 Falcon, et al., 2020). Adsorbing carboxymethyl cellulose (CMC) to the surfaces of whey 765 protein-coated rapeseed oil droplets was shown to reduce the rate and extent of lipid 766 digestion within in vitro and in vivo (rat feeding) studies (Malinauskyte, et al., 2018). 767 Studies have shown that coating fat droplets with particle-based emulsifiers (zein-PGA 768 nanoparticles) rather than conventional molecular-based emulsifiers (lactoferrin or 769 rhamnolipid) reduced lipid digestion and carotenoid bioaccessibility, which was 770 attributed to their ability to inhibit lipase accessing the emulsified lipid phase (Wei, et al., 771 2020). Similarly, other types of particle-based emulsifier have been shown to be effective 772 at inhibiting lipid or protein digestion in emulsions, including whey protein microgels 773 (Araiza-Calahorra, Wang, Boesch, Zhao, & Sarkar, 2020b; Sarkar, et al., 2016), whey 774 protein-dextran conjugate microgels (Araiza-Calahorra, Glover, Akhtar, & Sarkar, 2020a; 775 Araiza-Calahorra, et al., 2020b), cellulose nanofibers (Winuprasith, et al., 2018), and 776 cellulose nanocrystals (Sarkar, Zhang, Murray, Russell, & Boxal, 2017b). Numerous 777 other studies have shown that modulating the interfacial layers of fat droplets can alter 778 lipid bioaccessibility and/or nutraceutical bioaccessibility (Hu, Li, Decker, Xiao, &

McClements, 2011; Li, et al., 2010a; McClements, et al., 2010; Pinheiro, Coimbra, &
Vicente, 2016; Sarkar, Li, Cray, & Boxall, 2018). In particular, the importance of
molecular *versus* particulate and digestible *versus* indigestible interfacial coatings has
been stressed (Sarkar, Zhang, Holmes, & Ettelaie, 2019). Typically, indigestible
particulate coatings are more effective at inhibiting macronutrient digestion, provided
they remain attached to the macronutrient surfaces.

785 The digestion of macronutrients may also be inhibited when they are embedded 786 within hydrogel particles (Figure 6). For instance, the rate and extent of lipid digestion 787 has been shown to decrease when fat droplets are embedded within calcium alginate 788 microgels (Li, et al., 2011a). The degree of inhibition in lipid digestion increased as the 789 microgel dimensions increased, as well as when the pore size in the biopolymer network 790 decreased, which can be attributed to the longer distance the lipase molecules have to 791 travel to reach the entrapped fat droplets and the greater hindered diffusion, respectively. 792 Encapsulation of lipid droplets in other kinds of hydrogel-based microgels has also been 793 shown to inhibit their digestion, including those fabricated from carrageenan (Zhang, et 794 al., 2016), alginate-chitosan (Li & McClements, 2011b), gellan gum (Vilela, Perrechil, 795 Picone, Sato, & da Cunha, 2015) and egg white proteins (Gu, et al., 2017). Thus, the 796 lipid digestion profile can be modulated by controlling the type of hydrocolloids used to 797 create microgels, as well as the fabrication conditions, as this will affect how the 798 microgels response in different regions of the GIT (Figure 5) (McClements, 2017). 799 Encapsulation within microgels has also been utilized to reduce the digestion of other 800 macronutrients, such as proteins (Zhang, Zhang, & McClements, 2017) and starches 801 (Rose, et al., 2009). This approach may therefore be suitable for creating a new 802 generation of functional foods that can modulate the rate of macronutrient digestion, 803 thereby controlling blood metabolite levels and hormonal responses after food ingestion.

804 3

5.3.5. Gastrointestinal barrier properties

Some hydrocolloids alter the absorption of bioactive substances by changing the
permeability of the mucus layer or epithelium cells. For instance, chitosan has been
reported to act as a permeation enhancer for certain kinds of bioactive agents (Canali,
Pedrotti, Balsinde, Ibarra, & Correa, 2012), which has at least partly been attributed to its
ability to bind to and increase the dimensions of the tight junctions between epithelium

810 cells (Schipper, et al., 1997; Thanou, Verhoef, & Junginger, 2001)

811 5.3.6. Alteration of chemical stability of nutrients

812 In addition to their normal hydrolysis by digestive enzymes in the GIT, some 813 macronutrients may undergo other kinds of chemical changes in the gastrointestinal tract 814 that can impact their health effects. In particular, polyunsaturated lipids are prone to 815 oxidation under certain gastrointestinal conditions (Kerem, Chetrit, Shoseyov, & Regev-816 Shoshani, 2006; Larsson, Cavonius, Alminger, & Undeland, 2012; Nieva-Echevarria, 817 Goicoechea, & Guillen, 2020). As a result, they may form toxic reaction products that 818 can be absorbed by the human body and cause health problems (Goicoechea, Brandon, 819 Blokland, & Guillen, 2011). Some hydrocolloids (or their digestion products) are known 820 to have strong antioxidant properties (McClements & Decker, 2018; Sarmadi & Ismail, 821 2010; Teixeira, Pires, Nunes, & Batista, 2016), and so their presence within a food may 822 be able to inhibit the chemical degradation of polyunsaturated lipids in the GIT. Various 823 kinds of food proteins and polysaccharides have been shown to exhibit antioxidant effects 824 through different physicochemical mechanisms, including chelation of transition metal 825 ions and free radical scavenging effects (Laguerre, Lecomte, & Villeneuve, 2007; Nieva-826 Echevarria, et al., 2020).

827 **5.4. Modulation of Nutrient Bioavailability**

828 A widely explored application of hydrocolloids has been to alter the 829 pharmacokinetics (PK) and bioavailability of nutrients, *i.e.*, macronutrients, 830 micronutrients, and nutraceuticals. The PK profile of an ingested bioactive component 831 describes the change in its concentration in a specific tissue, often the systemic 832 circulation, over time (Figure 7). As discussed in Section 5.3, this information is 833 important because it influences the bodies hormonal and metabolic responses to ingested 834 macronutrients, which influences an individual's susceptibility to overeating, obesity, and 835 diabetes. The PK profile is also important because it determines the bioavailability of 836 nutrients. A number of key events contribute to the bioavailability of these bioactive 837 substances, which can be summarized by the following expression (Yao, Xiao, & 838 McClements, 2014): 839

$BA = B^* \times A^* \times D^* \times M^* \times E^*$ (17)

842	In this expression: B* represents <i>bioaccessibility</i> – the fraction of nutrient in the intestina
843	fluids in a form that can be absorbed; A* represents absorption – the fraction of the
844	bioaccessible nutrients that are actually absorbed by the body; D* represents distribution
845	- the fraction of the absorbed nutrients in the target tissues (often the bloodstream) after
846	distribution around the body; M^* is the <i>metabolism</i> – the fraction of nutrients in a
847	bioactive form after any chemical or biochemical reactions inside the human body
848	(before or after absorption); and, E^* is the <i>excretion</i> – the fraction of the bioactive
849	nutrient that has not been removed from the body, e.g., via the feces or urine. Typically,
850	one or more of these phenomena may limit the overall bioavailability of a nutrient,
851	depending on its molecular and physicochemical properties (McClements, Li, & Xiao,
852	2015). Each of these phenomena is time-dependent, which causes the nutrient levels in
853	the bloodstream or other tissues to vary over time. For instance, the nutrient levels in the
854	blood will typically increase sometime after ingestion of a food, reach a maximum level,
855	and then decrease as they are distributed, metabolized and excreted (Figure 7).
856	Food hydrocolloids may alter the PK profile of nutrients in a number of ways
857	(Boland, et al., 2014; McClements, et al., 2009), many of them discussed in detail in the
858	previous section:
859	• Some hydrocolloids (particularly proteins) have strong buffering capacities and
860	can therefore leader to a higher pH in the stomach after ingestion of a food, which
861	can alter the aggregation state of macronutrients and the activity of digestive
862	enzymes (such as gastric lipase and pepsin).
863	• Some hydrocolloids thicken or gel the gastrointestinal fluids, which can alter
864	mixing and mass transport processes.
865	• Some hydrocolloids alter the aggregation state of macronutrients, which
866	influences the surface area exposed to digestive enzymes. Hydrocolloids that
867	promote aggregation, tend to inhibit digestion and reduce bioavailability, whereas
868	those that prevent aggregation have the opposite effect.
869	• Some hydrocolloids form protective coatings around macronutrients, which
870	inhibits the ability of digestive enzymes from reaching them, thereby slowing

- 871 digestion and reducing bioaccessibility.
- 872 • Some hydrocolloids bind gastrointestinal components (such as bile salts, calcium, or enzymes), which can either promote or inhibit digestion and bioaccessibility. 873
- 874 • Some hydrocolloids have antioxidant properties and can therefore reduce the 875 chemical degradation of nutrients in the GIT.
- 876 877

Some hydrocolloids increase the permeability of the intestinal membrane, thereby • increasing absorption.

878 It is therefore possible to control the PK profiles of nutrients by controlling the type, 879 concentration, and structural organization of hydrocolloids in foods, which may be useful 880 for designing more effective functional foods to improve human health and wellbeing. 881 In some cases, the presence of hydrocolloids in foods may have beneficial effects on 882 nutrient absorption by increasing the bioavailability of oil-soluble vitamins or 883 nutraceuticals. In other cases, they may have the opposite effect. For instance, the ability 884 of chitosan to promote the precipitation of mixed micelles in the small intestine can 885 reduce the bioaccessibility of oil-soluble vitamins, such as vitamin D (Tan, et al., 2020). 886 In general, any dietary fiber that retards lipid digestion can also reduce the 887 bioaccessibility of encapsulated bioactive substances by decreasing the fraction released 888 from the oil droplets, as well as by reducing the amounts of mixed micelles formed to 889 solubilize them (Winuprasith, et al., 2018; Zhou, et al., 2020). Consequently, it is 890 important to carefully consider the overall impact of hydrocolloids on the gastrointestinal 891 fate of foods and nutrient bioavailability.

892

5.5. Modulation of Gut microflora

893 The complex community of microorganisms residing in the human colon, 894 collectively known as the gut microbiome, influences our susceptibility to chronic 895 diseases such as obesity, inflammatory bowel diseases, autoimmune diseases, diabetes, 896 atherosclerosis, and mental illnesses, and so plays a critical role in our health and 897 wellbeing (Albenberg & Wu, 2014; Chassaing, Vijay-Kumar, & Gewirtz, 2017; Dinan & 898 Cryan, 2017; Kataoka, 2016; Lloyd-Price, Abu-Ali, & Huttenhower, 2016; Tuddenham & 899 Sears, 2015). Fostering a healthy ecosystem within the human gut is therefore believed 900 to be critical for ensuring a healthy population. The bacteria living in the gut digest food

901 that is not digested and absorbed in the upper GIT, thereby extracting additional calories 902 that can be utilized as energy. They transform some of these food remnants into new 903 substances that may be advantageous to human health, such as vitamins, essential amino 904 acids, and short chain fatty acids. They can detoxify harmful substances in our foods, 905 such as the toxins found in some plant-based foods, by chemically transforming them. 906 They may also generate signaling molecules (such as hormones) that communicate with 907 the human body and regulate appetite, moods, and emotions. Finally, a healthy gut 908 microbiome can help to train and strengthen the human immune system, thereby leading 909 to improved overall health.

910 Hydrocolloids, such as polysaccharides and proteins, impact the type and number of 911 microbes residing within the gut microbiome (Cockburn & Koropatkin, 2016; Porter & 912 Martens, 2017; Yao, Muir, & Gibson, 2016). Consequently, there is considerable interest 913 from food, nutrition, and clinical scientists in controlling the gut microbiome by 914 supplementing the diet with beneficial hydrocolloids. Consuming high levels of 915 digestible carbohydrates (rapidly digestible starches) has an adverse effect on the gut 916 microbiome, whereas consuming high levels of non-digestible carbohydrates (dietary 917 fibers) has beneficial effects. Nevertheless, the effects of specific dietary fibers depend 918 on their precise molecular and physicochemical properties. One of the most important 919 attributes of fermentable dietary fibers is their ability to generate short chain fatty acids 920 (SCFAs) in the colon that act as an energy source, as well as produce regulatory 921 molecules that send signals to our bodies that modulate our metabolisms, reduce 922 inflammation, and communicate with our brains.

923 Dietary proteins may also have an impact on the human microbiome (Oliphant & 924 Allen-Vercoe, 2019; Yao, et al., 2016). Indeed, diets high in proteins and low in 925 carbohydrates were reported to have undesirable effects on the composition and function 926 of the gut microbiome (Yao, et al., 2016). Meat proteins contain relatively high levels of 927 L-carnitine, an amino acid that can be converted into trimethylamine N-oxide (TMAO) 928 by the gut microbes in the colon, which is a substance linked to coronary heart disease 929 (Zeisel & Warrier, 2017). On the other hand, plant proteins do not contain high levels of 930 L-carnitine and may therefore be better for heart health.

931 6. Hydrocolloids in Foods

932 Hydrocolloids may be incorporated into the human diet in a variety of ways. They 933 may simply be added as functional ingredients, such as thickening agents, gelling agents, 934 or stabilizers. Alternatively, they may be used to create complex structures in foods that alter the way nutrients behave, such as colloidal delivery systems. Finally, they may be 935 936 naturally present in whole foods, such as fruits, vegetables, cereals, nuts, or seeds, which 937 may be eaten raw or cooked, thereby altering their properties and behavior within the 938 human body. In this section, a brief overview of the impact of hydrocolloids in these 939 different kinds of application on the gastrointestinal fate of foods is given.

940 6.1. Hydrocolloids as Functional Ingredients

941 Hydrocolloids are often added to foods as thickeners, gelling agents, stabilizers, or 942 emulsifiers (Phillips, et al., 2009). In this case, they are usually being employed to obtain 943 some desirable techno-functional attribute in a food such as appearance, texture, shelf-944 life, or mouthfeel. Until recently, the potential of using the same functional ingredients to 945 modulate the behavior of foods inside the human body was not actively considered. 946 Nevertheless, there is great potential for using hydrocolloids as multipurpose functional 947 ingredients, to create desirable food and gastrointestinal effects. In this case, it is 948 important to select ingredients that have the desired techno-functional properties (such as 949 food texture and stability), as well as the desired nutritional properties (such as delayed 950 macronutrient absorption or increased nutraceutical bioavailability). In many cases, this 951 may require reformulation of existing products.

952

6.2. Hydrocolloid-based Delivery Systems

953 Hydrocolloids can be used to assemble complex colloidal structures in foods that can 954 be used to encapsulate, protect, and deliver bioactive components (Figure 8). These kinds 955 of delivery systems have been reviewed in detail elsewhere and so only a brief overview 956 will be given here (McClements, 2017; Shewan, et al., 2013).

957 • *Emulsions*: Oil-in-water emulsions or nanoemulsions can be formulated using 958 hydrocolloids as emulsifiers (McClements, et al., 2017). Hydrophobic nutrients 959 can be located inside the oil droplets, amphiphilic nutrients at the oil-water 960 interface, and hydrophilic nutrients in the water phase. Consequently, a

- 961 combination of different nutrients can be incorporated into a single delivery962 system.
- Biopolymer nanoparticles: Protein and/or polysaccharide nanoparticles that
 contain densely packed hydrocolloid molecules and little water can be formed
 using antisolvent precipitation methods (Joye & McClements, 2013).
 Nanoparticles assembled from hydrophobic proteins, such as zein or gliadin, are
 commonly used to encapsulate hydrophobic nutrients.
- 968 *Microgels*: Microgels are small particles that consist of a network of protein 969 and/or polysaccharide molecules and a relatively large amount of water (Zhang, et 970 al., 2017). They can be fabricated using many different approaches using gelling 971 hydrocolloids, including injection, templating, molding, and phase separation 972 methods. Hydrophilic nutrients can be trapped inside them, provided they are 973 sufficiently large and/or they stick to the molecules that make up the gel network. 974 Hydrophobic nutrients can be encapsulated inside oil droplets first, which are then 975 incorporated into the microgels. It is even possible to create microgel-in-microgel 976 systems to achieve special effects, such as improved protection or sequential 977 release of different components (Ma, Tu, Wang, Zhang, & McClements, 2018).
- 978

6.3. Hydrocolloids in whole foods

979 In many whole foods, the hydrocolloids are present in their natural environment, 980 such as intracellular or intercellular regions. The cell walls of edible plants consist of 981 complex hydrocolloid matrices comprised mainly of cellulose, hemicellulose, and pectin 982 (Holland, Ryden, Edwards, & Grundy, 2020). The presence of these cell walls alters 983 macronutrient digestion and phytochemical bioavailability by acting as steric barriers that 984 inhibit the ability of digestive fluids from coming into contact with the proteins, starches, 985 and lipids trapped inside the cells. When the cellular structures of plants are broken 986 down, the macronutrients are released making them more readily digestible. Moreover, 987 the phytochemicals may also be released from the cells making them more bioavailable 988 in the GIT. The disruption of the cell walls through food processing may therefore have 989 either detrimental or beneficial effects on animal and human nutrition depending on the 990 system. More rapid digestion of starch or fats may lead to spikes in blood sugar or lipid

levels, which can cause dysregulation of the metabolism and endocrinal systems, leading
to chronic diseases such as diabetes and obesity. Conversely, the enhanced release of
proteins or phytochemicals (such as carotenoids) may have beneficial effects due to their

994 health-promoting effects.

995 As an example, an *in vitro* study of the digestion of rice, showed that the uncooked 996 rice was less hydrolyzed than cooked rice, but that partially cooked rice (10 min) was 997 hydrolyzed the same as fully cooked rice (20 min) (Tamura, Singh, Kaur, & Ogawa, 998 2016). The authors suggested that the presence of the bran around the starch granules in 999 the uncooked rice were able to prevent the amylase from digesting the starch molecules. 1000 Other studies have also shown that starch digestion is inhibited by the presence of intact 1001 plant cell walls, as well as the degree of starch crystallinity, which depends on the degree 1002 of thermal processing (Li, et al., 2020). This suggests, that the location of the indigestible 1003 fibers relative to the macronutrient is important. In future, more work is required to 1004 understand the structural organization of plant-based foods, how they breakdown inside 1005 the human body, and how this impacts their nutritional attributes.

1006 7. Potential Health Benefits of Food Hydrocolloids

1007 Certain kinds of food hydrocolloids, particularly dietary fibers, are claimed to exhibit 1008 specific health benefits when consumed regularly at sufficiently high quantities. 1009 Recently, a comprehensive review and meta-analysis of the potential health benefits of 1010 dietary fibers was carried out (Reynolds, et al., 2019). The authors included data that was 1011 equivalent to 135 million person-years taken from 185 prospective studies and 58 clinical 1012 trials on 4635 adults. It was reported that there was a 15–30% reduction in deaths or 1013 incidences of a range of chronic diseases, including all-cause mortality, cardiovascular 1014 disease, coronary heart disease, stroke, type 2 diabetes, and colorectal cancer in 1015 observational studies that compared people consuming the highest levels of dietary fiber 1016 to those consuming the lowest levels. Moreover, people consuming high dietary fiber 1017 levels were also shown to have lower bodyweights, blood pressures, and cholesterol 1018 levels in clinical trials. The researchers reported that the greatest reduction in risk from 1019 chronic disease could be achieved by consuming around 25 to 29 g of dietary fiber a day, 1020 which is much higher than the levels consumed by the majority of people, in both

developed and developing countries. Interestingly, the authors only found a weak link
between overall disease risk and the glycemic index or glycemic load of a diet. Even so,
there was some evidence that diets with a low glycemic index or glycemic load did
reduce incidences of stroke and type 2 diabetes. Increased consumption of fiber-rich
fruits and vegetables has also been linked to a reduced risk of cardiovascular disease,
cancer and all-cause mortality in a systematic review and meta-analysis of the data
(Aune, et al., 2017).

1028 Overall, these studies suggest that it is important for consumers to eat more dietary 1029 fiber-rich foods to improve their health. Many of the health benefits reported for dietary 1030 fibers are related to the physicochemical and physiological processes that occur in the 1031 human gut after ingestion of food hydrocolloids discussed earlier, such as bile salt 1032 binding (cholesterol reduction), modulation of macronutrient digestion, fermentation in 1033 the colon, and alteration of stool rheology. The amount of dietary fibers in the diet can be 1034 increased by consuming more fruits, vegetables, grains, and nuts, however many people 1035 do not eat enough of these foods due to economic, social, or personal reasons. There are 1036 therefore opportunities to increase the level of dietary fibers in processed foods to 1037 enhance their healthiness, but there is some question about whether this approach will 1038 have the same health benefits. In other words, dietary fibers may have different effects 1039 inside the human gut when they are an integral part of natural foods (such as fruits, 1040 vegetables, or whole grains) than when they are isolated and added to processed foods as 1041 functional ingredients (Grundy, et al., 2016a; Grundy, Lapsley, & Ellis, 2016b; Guo, Ye, 1042 Bellissimo, Singh, & Rousseau, 2017b). This is an another important area for future 1043 research.

It should be noted that there may also be adverse effects associated with consuming
large quantities of certain dietary fibers, including bloating, gastrointestinal discomfort,
flatulence, and loose stools, especially in individuals with bowel disorders (Nyyssola,
Ellila, Nordlund, & Poutanen, 2020). These effects depend on the type and amount of
dietary fibers consumed, as well as the nature of the foods they are consumed with.
Consequently, both the health benefits and potential risks of fortifying foods with high
levels of dietary fibers should be carefully considered when formulating functional foods.

1051 **8. Conclusions**

1052 Hydrocolloids have been used as techno-functional ingredients in foods and beverages for decades, *e.g.*, as thickening, gelling, emulsifying, or stabilizing agents. 1053 1054 More recently, there has been a focus on their ability to modulate the processes occurring 1055 within the human gastrointestinal tract after the ingestion of foods. Researchers are 1056 attempting to develop structure-functional relationships for different hydrocolloids that 1057 link their molecular characteristics (such as molar mass, conformation, branching, charge, 1058 and polarity) to their ability to modulate specific gastrointestinal processes (such as mass 1059 transport, binding, solubilization, and absorption). As our understanding of these 1060 complex relationships increases it should be possible to design functional foods with 1061 specific health benefits, *e.g.*, the ability to increase nutraceutical or vitamin 1062 bioavailability, reduce fat or starch digestion, increase the satiety response, reduce 1063 cholesterol levels, decrease susceptibility to colonic diseases, or foster a healthy gut 1064 microbiome.

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Figure 1. The fraction of sites occupied on a receptor molecule increases as the lignd concentration and binding affinity increase.





Figure 2. Hydrocolloids increase fluid viscosity by perturbing fluid flow – the fluids have a longer distance to travel in the same time, which increases the friction and energy dissipation.



Figure 3. The ability of hydrocolloids to thicken solutions depends on their effective volume ratio, which depends on their molecular characteristics (molar mass, branching, conformation, and charge).



Figure 4: Hydrogel particles may breakdown in the GIT through a variety of physicochemical mechanisms, including shrinking, swelling, disintegration, erosion or fragmentation.

Figure 5: Bioactive agents can be released in different regions of the GIT by selecting biopolymers with different sensitivities to gastrointestinal conditions, such as pH, ionic strength, or enzyme activity.





Figure 6: Hydrocolloids may impact the aggregation state or interfacial properties of macronutrients, which alters their digestion and absorption.



Figure 7: The pharmacokinetic (PK) profile of a bioactive component (such as a vitamin or nutraceutical) represents the change in its concentration in a specific tissue (often the bloodstream) over time.



Figure 8: Examples of some kinds of colloidal delivery systems that can be prepared using hydrocolloids (not drawn to scale).

Table 1. Overview of some key molecular and functional attributes of common food-grade proteins used to as functional ingredientsin foods. Here pI is the isoelectric point, and T_m is the thermal transition temperature. Key: A: Type A Gelatin; B: Type B Gelatin; S:S-Type Ovalbumin; 7S and 11S Soy Glycinin fractions. As well as the traditional functional attributes mentioned here, many can alsobe used as structural components of colloidal delivery systems.

Name	Source	Main Structural Type	pI	~ T _m (°C)	Solubility	Functionality
β-lactoglobulin	Milk	Globular	~ 5.0	~75	Water	Emulsifying, gelling, and foaming
Caseins	Milk	Flexible	~ 4.6	~125-140	Water	Emulsifying, gelling, and foaming
Bovine Serum	Milk/Blood		Globular ~ 4.7	~80	Water	Emulsifying, gelling, and
Albumin		Globular				foaming
	Milk	Globular	~ 8.0	~60 & 90	Water	Emulsifying, gelling, and
Lactoferrin						foaming
		/hite Globular	~ 4.6	~74; 82 ^s	Water	Emulsifying, gelling, and
Ovalbumin	Egg White					foaming
_	Egg White		~ 11.0	~74	Water	Emulsifying, gelling, and
Lysozyme		Globular				foaming
	Egg Yolk	k Globular ~ 4	4.0	~80	Water	Emulsifying, gelling, foaming
Phosvitin			~ 4.0			and iron binding

Gelatin	Animal Collagen	Flexible	~ 8 ^A ~ 5 ^B	~ 5 (fish) ~ 40 (animal)	Water	Emulsifying, gelling, and foaming
Soy Glycinin	Soybean	Globular	~ 5.0	~67 ^{7S} ; 87 ^{11S}	Water	Emulsifying, gelling, and foaming
Zein	Corn	Globular	~ 6	~90	Organic Solvent	Nanoparticle formation
Pea globulins	Pea	Globular	~ 4.5	~72	Water	Emulsifying, gelling

Table 2. Summary of important molecular characteristics of some common food-grade polysaccharides used as functional ingredients

 in foods. As well as the traditional functional attributes mentioned here, many can also be used as structural components of colloidal

 delivery systems.

Name	Source	Main Structure Type	Major Monomer	Function
Carrageenan	Algal	Linear/Helical	Sulfated Galactan	Thickening, gelling (K ⁺ or Ca ²⁺), stabilizing
Xanthan Gum	Xanthomonas campestris exudate	Linear/Helical (High MW)	β-D-glucose (backbone)	Thickening, stabilizing, structure formation
Methyl Cellulose	Wood Pulp	Linear	Methylated Glucose	Thickening, stabilizing, gelling (thermoreversible)
Pectin	Plant Cell Walls	Highly Branched Coil	Glucuronate (backbone)	HM: Gelling (sugar + heat), stabilizing LM: Gelling (Ca ²⁺), stabilizing
Beet Pectin	Sugar Beet Pulp	Branched Coil with Protein	Glucuronate (backbone)	Emulsification, gelling (sugar + heat; Ca ²⁺ or laccase), stabilizing
Gum Arabic	Acacia Sap	Branched Coil Domains on Protein Scaffold	Galactose	Emulsification, film forming
Inulin	Plants or Bacteria	Linear with occasional branches	β-D-fructose	Prebiotic, thickening

Chitosan	Crustaceans, Invertebrates	Linear	2-amino-2- deoxy-β-D- glucose	Gelling (polyphosphate)
Alginate	Algal	Linear	β-D- Mannuronic Acid	Gelling (Ca ²⁺), stabilizing
Agar	Algal	Linear	β-D- Galactopyranose	Gelling (Ca ²⁺), stabilizing
Guar gum	Seeds	Linear with side chains	D-mannose and D-galactose	Thickening, stabilizing
Locust bean gum	Seeds	Linear with side chains	D-mannose and D-galactose	Thickening, stabilizing
Tara gum	Seeds	Linear with side chains	D-mannose and D-galactose	Thickening, stabilizing

Note: Commercially available polysaccharide ingredients typically contain appreciably different molecular and functional properties; the listed information

describes general characteristics for industrial usage.









Biopolymer Particles Biopolymer-coated Lipid Droplets

Microgels

Microgels-in-Microgels