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#### 1 Abstract

2 Gels possess remarkable properties, and they hold particular importance in food science. After consumption, food gels undergo large deformation, which impacts the overall texture of the 3 4 food. This process is influenced by various factors, including temperature, pressure, and presence of crosslinking agents. Comprehensive insights into the interplay among these factors 5 and gel texture, combined with the theoretical exploration of gel deformation, enable the 6 7 development of foods to meet consumer preferences. To bolster the development of food gels, in this review, we summarize the factors affecting the large deformation of gels. Moreover, we 8 9 discuss various mathematical models established by food scientists to explore the large deformation of food gels and explore applications thereof. We expect that these insights into 10 the large deformation of gels can lead to their increased utilization in the food industry. 11 12 Keywords: food gels; large deformation; theoretical basis; application

#### 13 **1. Introduction**

Amidst global sustainability challenges, particularly those arising from population growth, 14 environmental pressures, and energy consumption, development of innovative food products 15 that address human nutritional requirements has become increasingly critical (Kell, 2022; 16 Kumar et al., 2022). Among these innovations, gels have emerged as a promising solution. As 17 elastic, semisolid soft materials with a three-dimensional (3D) network structure, gels 18 19 demonstrate excellent controllability and large deformation characteristics (Gul et al., 2022; Yuan et al., 2025). The term "large deformation" indicates irreversible changes in the volume 20 21 and shape of a gel due to external stimuli such as temperature, pH, or mechanical load, resulting in the fracture or collapse of the gel matrix and the permanent disruption of its molecular 22 network. This property not only determines the performance of gels during food processing but 23 also considerably affects the texture of food products (Gheorghita Puscaselu et al., 2020). Gels 24 are used in a wide range of foods, from desserts, yogurts, to coatings for baked goods and fruits 25 (Silva et al., 2020; Górska et al., 2024). In addition, they can be used as fat substitutes to 26 maintain the nutritional value of foods and reduce their caloric content, thereby decreasing the 27 risk of chronic conditions such as cardiovascular diseases and obesity (Nath et al., 2023; 28 Nepovinnykh et al., 2019; Yang et al., 2020). However, the large deformation properties of gels, 29 which are essential for their structural and functional optimization, remain poorly understood. 30 In laboratory settings, research on gel deformation primarily focuses on texture and 31 32 rheological properties of gels. Texture analysis involves the assessment of various characteristics, such as stretching, compression, and puncture (St. Pierre et al., 2024), while 33 rheological analysis primarily focuses on the viscoelastic properties and stress-strain responses 34 of gels (Y. Wang & Selomulya, 2022). Modern rheometers can precisely determine the manner 35 in which complex materials react to applied stress or strain, enabling the characterization of 36 gel composition, structure, and processing effects. Customized gels with required 37

characteristics can be fabricated by manipulating the polymer microstructure and surrounding 38 medium (Nath et al., 2023). The complete rheological deformation curve of a viscoelastic gel 39 40 typically comprises linear, nonlinear, and fracture regions (Fig. 1). Deformation is minimal in the linear region, whereas the nonlinear region shows more substantial deformation and is thus 41 termed the "large deformation" region (Zhang et al., 2007). Under industrial processing 42 conditions, gels are exposed to high levels of applied stress and strain. In such cases, mimetic 43 44 mechanical tests prove to be invaluable for rheological characterization (Walayat et al., 2022). In a previous review, Sinha et al. (2024) discussed a novel bi-gel system that combined 45 46 the properties of organogels and hydrogels, focusing on how adjusting the organogel and hydrogel fractions can regulate their properties. Meanwhile, Lin et al. (2020) comprehensively 47 reviewed the preparation methods for different types of emulsion gels (including bulk emulsion 48 gels, emulsion gel particles, and fluid emulsion gels), their structure-property relationships, 49 and their applications in the food industry. Moreover, B. Liu et al. (2024) systematically 50 classified the mechanisms of food gel synthesis and discussed the application potential of gels 51 in various functional foods, highlighting their applications in food packaging, satiety gel 52 systems, nutrient delivery systems, food coloring adsorption, and food safety monitoring. 53 While considerable research has been conducted on gels in food science, there remains a critical 54 gap in understanding the manner in which the large deformation properties of these gels can be 55 optimized for diverse applications. Although the existing research predominantly focuses on 56 isolated factors such as pH, temperature, and crosslinking mechanisms, the interplay among 57 these factors remains underexplored. Moreover, advancements in theoretical modeling and 58 experimental tools have yet to be fully integrated into practical applications, limiting their 59 potential effect on food innovation. Therefore, this review examines the factors influencing the 60 large deformation of gels and discusses the mathematical models established by food scientists 61 to explore this deformation in the context of food gel performance. This review aims to 62

synthesize the current knowledge regarding the large deformation properties of gels, highlight
the research gaps, and propose future directions to bridge the divide between fundamental
research and industrial applications.

#### 66 2. Theoretical framework for the large deformation of gels

The large deformation behavior of gels involves two key phenomena: (1) nonlinear elastic 67 response, in which the relationship between force and deformation becomes nonlinear, and (2) 68 69 material failure at a critical strain, referred to as the breaking strain. Both these conditions can be studied using stress-strain curves (Bot et al., 1996). Damage and fracture both need to be 70 71 modeled to accurately simulate gel behavior. However, one key challenge lies in understanding the effect of fluid content on gel fracture. In addition, many models underestimate fracture 72 resistance because they ignore the energy dissipation caused by fluid diffusion (Lake & Thomas, 73 1967). To address these issues, theoretical models and simulations need to incorporate fluid 74 diffusion alongside the large deformation, damage, and fracture mechanisms that characterize 75 gel behavior. 76

Mao and Anand (2018) developed a theoretical framework to explain the fracture of polymeric gels, incorporating the combined effects of fluid diffusion, large deformation, and damage. Their theory introduced two key ideas. First, it accounted for the changes in a gel's free energy due to entropy and internal-energy shifts caused by the stretching of polymer chains. Second, it posited that gel damage occurs through chain-scission, in which polymer chains break because of internal-energy changes rather than configurational entropy changes.

83 Small molecules can migrate into polymer networks, causing the network to swell and 84 form polymeric gels. Hong et al. (2008) developed a theory combining mass transport and large 85 deformation to explain this behavior. This theory suggested that the free energy of the gel arises 86 from two molecular processes: the stretching of the polymer network and its mixing with small 87 molecules. Small molecules and polymer chains both were treated as incompressible, and

osmotic pressure was modeled using a Lagrange multiplier. The theory demonstrated that gels 88 can deform through (1) rapid local rearrangement of molecules, which changes their shape 89 without altering their volume, or (2) slower migration of small molecules, which affects both 90 shape and volume. These mechanisms were modeled assuming that small molecules diffuse 91 92 within the gel. This theory explains gel behavior under various conditions, such as those when a constrained gel layer is subjected to weight or deformation by means of a conical indenter. 93 94 Such advancements enable more accurate modeling and a deeper understanding of gel deformation in complex scenarios. 95

#### 96 2.1. Theoretical basis of the large deformation of gels

97 2.1.1. Free energy functions

Theories for hydrogel deformation were first proposed in pioneering works by Flory and 98 Rehner (1943). These initial theories suggested that the Helmholtz free energy of a polymer 99 100 network in an aqueous solution can be represented as the additive decomposition of the free energy of the elastic stretch of the polymer network and the free energy of mixing when the 101 polymer network interacts with a solution. This decomposition has been widely used in 102 subsequent theories of hydrogel behavior. Yang and Wang (2019) developed governing 103 equations for functional gradient spherical hydrogels under spherically symmetric conditions 104 by using the Flory-Huggins free energy function to simulate the nonuniform and large 105 deformation swelling behavior of these hydrogels at a given internal pressure and chemical 106 107 potential. According to the Flory theory, the more general form of the free energy of a hydrogel can be described as follows: 108

109

$$W = W_{net} + W_{mix} \sum_{r \neq s} W_r \tag{1}$$

110 where  $W_{net}$  and  $W_{mix}$  denote the free energy of network stretch and that of mixing, respectively. 111 These two entities are applicable to all types of hydrogels. Meanwhile,  $W_r$  represents the free 112 energy contributions of factors other than the solvent and is specific to certain types of gels.

These free energies include, but are not limited to, free energy of ionization (Wion), polarization 113 (W<sub>pol</sub>), and dissociation (W<sub>dis</sub>), which have been detailed previously (Z. Liu et al., 2015) and 114 thus have not been repeated in this review.

2.1.2. Coupled diffusion-deformation theory 116

Baek and Srinivasa (2004) established thermodynamic theories of coupled diffusion and 117 large deformation, paving the way for future continuum mechanics studies of hydrogel swelling. 118 119 Yang et al. (2022) investigated the transient behavior of diffusion and deformation in hydrogels on the basis of the coupled diffusion theory. They stated that the diffusion of solutes from a 120 121 spherical hydrogel occurs under the influence of fluctuating stresses. Meanwhile, the corners of a square hydrogel swell faster than the edges and interior, leading to deformation. Recent 122 developments in coupled diffusion-deformation models have addressed some of these gaps by 123 integrating solvent movement with mechanical deformation. However, these models often 124 require extensive computational resources and are yet to be experimentally validated in 125 complex food systems. Moreover, most existing models assume uniform material properties, 126 limiting their accuracy in describing hybrid gels or multicomponent systems, both of which are 127 commonly used in food formulations. 128

The equations governing gel swelling depend on the migration of solvent molecules. The 129 flux is expressed (Bouklas & Huang, 2012) as follows: 130

- $j_i = \frac{cD}{kT} \frac{\partial \mu}{\partial x_i}$ (2)131
- 132

115

Moreover, it follows conservation laws:

133 
$$\int_{V} \frac{1}{\det F} \frac{\partial C}{\partial t} dV + \int_{A} j_{i} dA = 0$$
(3)

where K is the Boltzmann constant, T is the temperature, V is the reaction volume, A is the 134 reaction area, t is the time, C is the solvent concentration, D is the constant of solvent diffusivity, 135 F is the deformation gradient,  $\mu$  is the chemical potential, and c is the true solvent concentration. 136 2.1.3. Theories for large deformation of hydrogels 137

Under the influence of external stimuli, a hydrogel undergoes deformation from its initial reference state to its final current state. In describing this process of deformation, the deformation gradient  $F_{iK}$  (or F) is conventionally used. Fig. 2 shows the state of deformation of a hydrogel. Dutta et al. (2020) examined the hygroscopicity of white rice using deformation gradient equation modeling to identify a set of optimal process parameters in a temperature range of 25°C–80°C.

In the current state at time t, the marker X moves to a place with coordinates x(X,t). The deformation gradient is denoted (Z. Liu et al., 2015) as follows:

146  $F_{iK}(X,t) = \frac{\partial x_i(X,t)}{\partial X_K}$ (4)

Under conditions of equilibrium, the change in the free energy of the hydrogel is balanced via the external work performed on the gel. This thermodynamic equilibrium is usually expressed (Toh et al., 2014) as follows:

150  $\int_{V} \delta W \, dV = \int_{V} B_{i} \delta x_{i} dV + \int_{A} T_{i} \delta x_{i} dA + \sum \left( \mu^{r} \int_{V} \delta C^{r} dV \right) \tag{5}$ 

where W is the free energy of the gel;  $B_i$  is the external force;  $T_i$  is traction; and  $\mu^r$  and  $C^r$  are the chemical potential and concentration of the r species, respectively.

#### 153 2.2. Numerical simulation of hydrogel behavior

Numerical simulation is often employed to predict the behavior of hydrogels because it allows for us to study gels with complex geometries, which are often excessively complicated to study via analytical methods. The common simulation tools employed to study hydrogel behaviors include the finite element method, meshless methods, and molecular dynamics simulation.

159 2.2.1. Finite element method

160 The finite element method is a numerical solution method used for solving problems in 161 elastic mechanics (Y. Liu et al., 2021). This method was rapidly developed following the 162 availability of the electronic computer. Gels are soft materials and can undergo large

deformations, and their mechanical behavior is influenced both by their polymer and solvent 163 compositions. Currently, experimental methods are insufficient to accurately determine the 164 mechanical properties of gel materials. Therefore, the finite element method has emerged as an 165 important and commonly used simulation tool for study of gel deformation. With the 166 development of monophase theories, the finite element simulation of hydrogel swelling has 167 been expedited even further. ABAQUS and COMSOL Multiphysics, two finite element 168 169 software, are currently the most popular tools employed for these simulations. ABAQUS can define material models through user-defined subroutines, while COMSOL Multiphysics is 170 171 often used for the simulation of heavily coupled physical material models (Wu et al., 2023).

Nakauma et al. (2014) used a finite element model to simulate the compression of agar 172 gel on different substrates, using linear elastic properties for their analysis. Their simulation 173 results were validated through experimental findings. Meanwhile, Y. Liu et al. (2015) 174 developed a finite element algorithm to model the uneven swelling of neutral hydrogels at 175 equilibrium. In addition, Toh et al. (2014) employed the finite element method to simulate large, 176 uneven deformations in gels under geometric constraints. Interestingly, using a custom 177 subroutine in ABAQUS, they modeled gels as hyperplastic materials. These studies showed 178 that finite element models can accurately predict the deformation of photothermal gels with 179 complex shapes. 180

181 2.2.2. Ogden model

182 The Ogden model (Bergström, 2015) is a very general mathematical hyperelasticity model 183 in which the Helmholtz free energy per reference volume is expressed in terms of the applied 184 principal stretch. The Ogden model can be used to describe the mechanical behavior of 185 materials under conditions of large deformation. This model is particularly suitable for 186 hyperelastic materials, such as rubber and some polymers, as these materials exhibit nonlinear 187 stress–strain relationships under the influence of large strains. 188 The first-order Ogden model expresses the mathematical relationship of compression 189 stress with gel concentration, applied to fit the experimental data (Yao et al., 2022):

190 
$$\sigma = \frac{2\mu}{\alpha} \left( \lambda^{\alpha} - \lambda^{\left(-\frac{\alpha}{2}\right)} \right)$$
(6)

191 where  $\sigma$  is the shear stress,  $\mu$  is the shear modulus, and  $\alpha$  is the Ogden constant.

The Ogden constitutive model has been widely adopted to describe the hyperelastic behavior of soft materials such as hydrogels and living tissues in the presence of uniaxial compression (Nedrelow et al., 2023). The first-order Ogden model was observed to be wellfitted to the effect of carbohydrates on the large deformation behavior of fish skin gelatin (X. Li et al., 2020). Moreover, Czerner et al. (2016) used the Ogden model to study the deformation and fracture behavior of physical gelatin gel systems and determined the shear modulus and strain hardening capability of the gels by means of the stress–stretch ratio curve.

199 2.2.3. BST model

In 1974, Blatz et al. (1974) proposed a strain energy density function based on a generalized measure of strain, which was later employed to study the large deformation of gels. This framework is referred to as the BST model. The BST model was derived from the rubber elasticity theory and has been employed to characterize the curvature of the stress–strain profile beyond the linear range. This model has been successfully applied to describe the deformation mechanics of various biopolymer food gels, including alginate (Zhang et al., 2005), mixed soy and κ-carrageenan (Cavallieri et al., 2010), and gelatin gels (Gravelle & Marangoni, 2021):

207 
$$\sigma = \frac{2E}{3n} (\lambda^n - \lambda^{-2n}) \qquad n \sim \frac{d_f}{d_{f^{-1}}}$$
(7)

208 where  $\sigma$  is the shear stress,  $\lambda$  is the stretch ratio, E is Young's modulus, and n is an elasticity 209 parameter.

210 Theoretical models such as the Ogden and BST frameworks have significantly advanced 211 the understanding of gel deformation, particularly in terms of comprehending nonlinear elasticity and fracture behavior. However, their application in food systems remains limited
because of several challenges. The Ogden model, while effective at describing nonlinear stress–
strain relationships, does not account for time-dependent viscoelastic properties, which are
critical in dynamic processes such as extrusion and chewing. Similarly, the BST model offers
robust predictions of gel fracture but struggles to incorporate solvent migration and
environmental factors such as pH and temperature variations.

218 Thus, future advancements should focus on integrating these theoretical frameworks with machine learning and experimental data so as to improve their predictive power and practical 219 220 relevance. For instance, machine learning could be employed to identify patterns in large datasets generated in rheological experiments, enabling models to better comprehend the 221 multiscale behavior of gels. Moreover, hybrid models that combine microstructural dynamics 222 with macroscopic deformation behavior might provide a more comprehensive understanding 223 of gel mechanics under real-world conditions. By addressing these limitations, theoretical 224 models might play a pivotal role in optimizing gel properties for applications such as 3D food 225 printing, the development of dysphagia-friendly formulations, and bionic food design, bridging 226 the gap between fundamental research and industrial practice. 227

#### 228 **3. Factors affecting the large deformation of gels**

Various factors can influence the large deformation behavior of gels and can be split into 229 two groups: internal and external. Notably, the internal factors are intrinsic to the gel and 230 231 include gel concentration and gel composition. Meanwhile, the external factors include pH, temperature, high-pressure processing (HPP), crystal addition, crosslinking agents, and 232 polysaccharide charge density. Food texture is affected by the large deformation characteristics 233 of food gels (Gravelle & Marangoni, 2021). Understanding the manner in which these factors 234 influence food properties might aid the development of novel food products customized to 235 consumer demands. 236

#### 237 3.1. Gel composition

Gel composition is an important intrinsic factor affecting the mechanical strength of gels. Proteins and polysaccharides are the most common gel components used in food products and have attracted considerable attention in the field of functional foods because of their low toxicity, edibility, biocompatibility, biodegradability, and affordability (K. Liu et al., 2021).

Polysaccharides strengthen gels by enhancing hydrogen bonding and forming robust double-network structures. For example, viscoelastic analysis has shown that curdlan expedites the formation of denser rice starch gels with a higher degree of solidity (Wang et al., 2024). Similarly, *Mesona chinensis* polysaccharide–tapioca starch gels exhibit excellent flexibility and durability, with a stable network structure maintained via hydrogen bonding (Huang et al., 2023).

Gelatin, a type of protein, facilitates the formation of hydrophobic interactions, disulfide 248 bonds, and hydrogen bonds among globular proteins, transforming weak 3D gel networks into 249 dense-layered structures. Gelatin can be categorized as aquatic or terrestrial depending on its 250 source. In particular, aquatic gelatin is primarily derived from fish (fish skin, fish scales, and 251 swim bladders), while terrestrial one is usually derived from the skin, bones, and connective 252 tissues of swine and cattle (Derkach et al., 2022; Nurilmala et al., 2021; Zou et al., 2024). 253 Among the different types of gelatin extracted from animal skins (pig skin, cow skin, and fish 254 skin), pig skin and cow skin gelatin exhibit the greatest and poorest ability to resist deformation, 255 respectively (Michelini et al., 2020). The gel strength of bovine bone gelatin, however, is 256 greater than that of fish skin gelatin (T. Zhang et al., 2020). These variations highlight the 257 manner in which the source and amino-acid composition of gelatin affect its ability to resist 258 large deformations. The energy storage modulus of globular protein gels significantly increases 259 when gelatin is added, suggesting that gelatin can improve the strength of these gels and 260 stabilize the protein network structure (Ma et al., 2022). 261

The choice of gel components—proteins or polysaccharides—greatly influences the mechanical and sensory properties of a gel. While polysaccharides facilitate robust network formation via hydrogen bonding, proteins provide a tunable rheological behavior. Previous research has largely focused on these components in isolation, neglecting the manner in which hybrid gels comprising proteins and polysaccharides may offer superior large deformation characteristics. Thus, comparative studies on hybrid gel systems could facilitate the development of novel gel formulations for customized applications.

#### 269 *3.2. Gel concentration*

270 Gel concentration is another intrinsic factor affecting the mechanical strength of gels. The minimum protein concentration required to form gels varies from one protein to another. For 271 example, the minimal concentration required for gel formation is 0.6% for gelatin, 3% for egg 272 273 albumin, 6.6% for soy proteins, and 4%-12% for whey proteins and varies depending on the pH and ionic strength (Le et al., 2017). In particular, gel concentration affects gel strength by 274 altering the gel's microstructure. In low-concentration gels, the microstructure is loose and the 275 pore size is large. Meanwhile, high-concentration gels are dense and have a small pore size 276 (Cortez-Trejo et al., 2021). Horinaka et al. (2022) studied the effect of gel concentration on the 277 cyclic deformation behavior of  $\kappa$ -carrageenan hydrogels. High-concentration  $\kappa$ -carrageenan 278 hydrogels exhibited good elastic properties, whereas low-concentration κ-carrageenan ones 279 showed a higher Young's modulus. Subsequently, Horinaka and Ogawa (2023) also explored 280 281 the effect of agarose concentration on the cyclic deformation behavior of agarose hydrogels, and their findings were consistent with those related to  $\kappa$ -carrageenan hydrogels. 282

Gel concentration considerably impacts the mechanical properties and deformation behavior of gels. However, while high-concentration gels often exhibit improved strength and reduced deformability, the manner in which these properties interact with other factors—such as crosslinking density or temperature—remains poorly understood. Future studies should explore synergistic effects by investigating the manner in which the optimization of gelconcentration can enhance gel stability during industrial processing conditions.

289 *3.3. pH* 

The effects of pH on the large deformation of gels are primarily mediated through pH's 290 effect on gel swelling, structural stability, and microstructure (Z. Liu et al., 2024). The response 291 of different gels to pH varies based on factors such as gel composition, crosslinking agents, 292 293 and environmental conditions. pH influences the structure of proteins by altering the balance of attractive and repulsive forces, which can lead to protein unfolding and denaturation (Tan et 294 295 al., 2021). For example, Razi et al. (2019) studied egg white albumin and basil seed gum mixtures under different pH conditions at high pressure. The gels formed at pH 5.0 exhibited 296 higher elasticity than those formed at pH 7.0, likely because of stronger electrostatic and 297 hydrophobic interactions at pH 5.0 (Hosseini-Parvar et al., 2016). These findings showed that 298 pH is a critical factor influencing the large deformation behavior of gels, highlighting its role 299 in shaping of gel properties. 300

Proteins, as natural polyampholytes, can form gels through heat-induced gelation or cold 301 gelation (Betz et al., 2012). The pH-dependent swelling behavior of protein gels has been 302 extensively studied. Saglam et al. (2013) observed that protein particles in gels formed via 303 emulsification and heat-induced gelation remained stable across a wide pH range but exhibited 304 aggregation at pH ~5 likely because of reduced electrostatic repulsion near the protein's 305 isoelectric point. This aggregation can cause substantial volume changes, affecting the 306 rheological properties of the gel. Wang et al. (2021) studied whey protein gels containing 307 glucose and fructose fabricated via the Maillard reaction at different pH levels. They observed 308 that the elastic modulus of these gels depended on the strength of interactions between the 309 protein molecules and sugar chains. The gels reached their maximum elasticity at different 310 temperatures, and their elasticity decreased as the pH increased, demonstrating that pH 311

312 influences molecular interactions within gels.

#### 313 *3.4. Temperature*

314 The treatment temperature of gels is crucial for determining their strength and rheological properties. Meat, a common component of the diet for several populations worldwide, exhibits 315 gel-forming and viscoelastic properties that depend on the bonds stabilizing the gel network 316 (Tammatinna et al., 2007). Changes in treatment temperature can affect the strength, rheology, 317 318 and sensory attributes of meat gels. For example, Mad-Ali et al. (2018) observed marked differences in gel strength and deformation among goat meat gels treated at different 319 320 temperatures. They noted that preheating the meat prior to heating it to 90°C could enhance the gel's breaking force and deformation capacity, highlighting the importance of temperature in 321 controlling meat gel behavior. 322

Starches too are key components of processed foods, and the gelatinization process during 323 heating affects the rheological behavior of starch gels (Rocha et al., 2011). Albano et al. (2014) 324 studied gels made from Peruvian carrot starch at different temperatures (10°C, 30°C, 50°C, and 325 70°C) and observed that they exhibited shear-thinning behavior and high thixotropy. Notably, 326 the results demonstrated that the setting temperature substantially affected the consistency 327 index of the gels. Further, Sha et al. (2019) observed that higher extraction temperatures of 328 porcine gelatin reduced the gel strength, gelation point, and melting point, demonstrating the 329 manner in which temperature affects the gelling properties of gelatin. 330

While pH and temperature are known to affect swelling and gelation kinetics, their interaction with mechanical stress during large deformation remains underexplored. For instance, acidic conditions can increase elasticity in egg albumin gels; however, the manner in which these conditions interact with thermal cycles to influence fracture behavior is still unclear.

335 *3.5. High-pressure processing* 



6 HPP, a nonthermal sterilization method in which pressures ranges from 100 to 1000 MPa,

is applied to food products, and it prevents spoilage without affecting the sensory or
physicochemical characteristics of food (Fam et al., 2020; C. Ren et al., 2020). This treatment
also provides functional effects and is widely used in the food industry (Mokrushin et al., 2022).
HPP can lower the gelatinization temperature of starch, with higher pressures providing greater
reductions in the gelatinization temperature (Balakrishna & Farid, 2020). In addition, HPP
treatment can be employed to obtain starch gels with more stable crystallites, denser structures,
reduced retrogradation, and increased gel stability (Guo et al., 2022).

Food products often comprise proteins and polysaccharides, which affect the texture, 344 345 structure, and stability of gels because of their gelling, thickening, and surfactant properties. In mixed gels, HPP disrupts the interactions between proteins and polysaccharides, causing 346 biopolymer complexes to dissociate and proteins to denature, which alters gel properties (Razi 347 et al., 2019). Previously, HPP has been successfully applied in fish processing (Qin et al., 2022). 348 Moreover, this technique is employed to modify protein properties in several foods, altering 349 protein structure and affecting coagulation, aggregation, and gelation behavior (Moreno et al., 350 2015). 351

Velazquez et al. (2021) showed that HPP can create a soft flexible network in crabmeat gels, reducing their rigidity by fully denaturing the proteins and facilitating aggregation through disulfide bonding. In particular, HPP at 100 MPa afforded the crabmeat gels with optimal rheological properties. Similarly, Xu et al. (2024) observed that HPP caused the gradual unfolding of  $\alpha$ -helixes in Tai Lake whitebait myofibrillar protein gels, transforming the  $\alpha$ helixes into  $\beta$ -sheets. Compared with untreated gels, these gels exhibited a 4.8-fold increase in gel strength and improved elastic and viscous moduli (G' and G''').

359 *3.6. Crystals* 

360 The large deformation properties of gels are influenced by natural crystallization during 361 aging and can also be affected by the addition of external crystals. Aging causes retrogradation in gels, gradually altering their physical properties (Jiang et al., 2020). This process involves
the crystallization of molecules in the gel, which results in the formation of an ordered structure.
Retrogradation affects not only the texture, rheological behavior, and pasting properties of gels
but also their crystalline and molecular structures (X. Liu et al., 2021).

Inulin, an oligosaccharide, forms gels containing a network of small crystals when mixed with water at high concentrations (J. Xu & Kenar, 2024). With time, these gels develop larger crystals and their fracture strain decreases. The retrogradation temperature also plays a role in this process: lower temperatures make the gels more brittle, and the maximum fracture force is generated during large deformation. Starch gel retrogradation is influenced by the water content, with higher water levels facilitating the formation of softer gels with well-defined crystallites (X. Liu et al., 2021).

The addition of crystals also affects gel deformation. Xiao et al. (2020) added cellulose nanocrystals (CNCs) to whey protein gels, increasing their water-holding capacity, strength, viscoelasticity, and thermal stability. CNCs restricted the water mobility within these gels and transformed the  $\alpha$ -helixes of proteins into  $\beta$ -sheets. These findings were similar to those reported by X. Liu et al. (2021) who highlighted the role of moisture in gel deformation. Thus, naturally sourced CNCs are being explored as promising gel modifiers in the food industry.

379 *3.7. Crosslinking agents* 

The water absorption capacity of a hydrogel depends on factors such as the type and density of the polymer and crosslinking agent used (Juan et al., 2022). Crosslinking agents are chemicals that link unsaturated compounds at the molecular level, forming a 3D network structure in gels. These agents are used in various fields, including the food industry, biomedicine, and materials science. In the food industry, enzymes and certain chemicals are the most commonly employed crosslinking agents (Table 1). Enzymatic crosslinking agents such as transglutaminase (TG) and various oxidases catalyze the formation of covalent bonds among proteins (Zheng et al., 2022). Chemical crosslinkers are typically small molecules that interact with proteins or other food components to yield a stable crosslinked framework, thus influencing food characteristics, particularly food texture. Other than these conventional crosslinkers, some specialized crosslinking agents can also be employed for specific applications. For instance, a novel crosslinking agent that contains whey protein, papain, glycerol, and epicatechin gallate exemplifies the recent advancements in crosslinking strategies (Liu et al., 2023).

Gelatin, a biopolymer derived from collagen, is widely used in food and pharmaceuticals owing to its swelling capacity, biodegradability, and biocompatibility (Echave et al., 2019). Although gelatin can form hydrogels upon cooling, its mechanical properties are often poor, necessitating the use of crosslinking agents (Uranga et al., 2020). Lin et al. (2023) observed that TG can crosslink fish gelatin (FG) in a dose-dependent manner, notably increasing gel strength (G'). Thus, the introduction of covalent crosslinks via TG can enhance the thermodynamic and mechanical stabilities of FG gels, creating more organized structures.

In summary, crosslinking agents greatly improve the strength, stiffness, and stability of gelatin hydrogels, which makes them valuable across various industrial sectors. However, the influence of crosslinking agents on the large deformation of gels warrants further exploration as this could help in the development of gels with improved properties and provide insights into the internal factors governing their large deformation.

Crosslinker	Source	Polymers	Mechanism	pH,
				Temperature
Transglutami	Bacteria,	Gelatin, collagen,	Presence of glutamic acid and	5.5, 40°C
nase	mammals,	whey protein, soy	lysine groups; acyl	
	and plants	protein, chitosan,	ligands/amine ligands from L-	
		and carboxymethyl	glutamic acid and L-lysine	
		cellulose		
Horseradish	Root	Hyaluronic-	Oxidation of phenolic and	7.0, 45°C
peroxidase	(Armoracia	Tyramine	catechol groups (requires	
	rusticana)	(®Corgel),	hydrogen peroxide addition),	
		alginate-tyramine,	peroxidation, isomerization,	
		gelatin	dimerization, and enolization	
		norbornene/4-arm		
		poly(ethylene		
		glycol) dihydrogen		
		tetrazine, and silk-		
		fibroin		
Tyrosinase	Microorganis	Alginate,	Presence of phenolic/catechol	6.5, 50°C
	m,	hyaluronic	groups. Oxidation (O <sub>2</sub>	
	plants, and	acid/gelatin, and	species), semi-quinone	
	animals	glycol chitosan	formation through Micheal	
			addition, isomerization,	
			dimerization, and enolization	
Whey	Animals	Protein and	Maillard reaction	7.0, 50°C
protein		polysaccharides		
Epicatechin	Plants	Gelatin	Aldehyde group in gallic acid	5.5, 45°C
gallate			reacts with the amino group to	
			form a stable covalent bond	

406 <b>Table 1.</b> Crosslinking agents used in common gels (Al	avarse et al., 2022).
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Although considerable efforts have been made toward understanding the specific

408 mechanisms through which these substances impact the large deformation in gels, continued 409 research is still needed. Some researchers are now employing computational mechanics to 410 examine the large deformation of gels, providing theoretical information that could aid the 411 understanding of and improvement in gel properties.

412

#### 4. Applications of the large deformation of gels

Gels, as abundant renewable biopolymers, have attracted attention because of their 413 414 sustainability, affordability, and biocompatibility. Consequently, they have emerged as useful, cost-effective, and easily modifiable materials (Li et al., 2021). Their unique properties and 415 416 wide application prospects have sparked interest across various sectors, including food (De Leon Rodriguez & Hemar, 2020), biomedicine (Z. Li et al., 2020), soft robotics (Jiang et al., 417 2023), and flexible electronics (Zhu et al., 2024). Gels have widespread applications in food 418 products (Fig. 3), and gel texture notably impacts the taste of foods. Exploring the large 419 deformation characteristics of gels, such as gel strength and viscoelasticity, can help in the 420 development of foods customized to consumer preferences. Moreover, gels can be used for the 421 production of easy-to-consume foods for individuals with chewing difficulties, thus addressing 422 their specific needs. 423

#### 424 *4.1. Food texture*

Texture refers to the sensations experienced when eating food, including the initial bite, 425 chewing, and swallowing (Kutlu et al., 2022). Commonly referred to as "mouthfeel," texture 426 plays a key role in consumer perception because it represents the sensory experience of a food's 427 molecular, microstructural, and macrostructural composition. Therefore, texture is evaluated 428 through visual assessment, touch, and sound (Cao & Mezzenga, 2020). Among these factors, 429 the tactile feedback provided by the mouth during eating is the most important. The first bite, 430 which breaks down the structure of the food, providing different sensory perceptions based on 431 the gel's fracture stress and strain (Franks et al., 2020). Gels with higher deformation capacity 432

433 provide a softer initial bite but remain resilient, providing a satisfying chew. The large 434 deformation behavior of gels also impacts the manner in which the gel breaks down and 435 releases flavors, which is a crucial consideration while designing foods to suit consumer 436 preferences. For example, softer gels are preferred in desserts, while firmer, more elastic gels 437 are used in products such as gummies or meat analogs for a chewier texture.

The texture of solid and semisolid foods is attributed to the mechanics of food breakdown in the mouth (van Bommel et al., 2019). The consumer's first bite and chew are governed by the large deformation properties of the food (Gravelle & Marangoni, 2021). Instruments can be employed to analyze the fracture properties of gels, such as fracture stress and strain, which are directly related to their texture. In particular, fracture stress is associated with the perceived hardness of foods, while fracture strain is related to how easily the food deforms during the first bite (Nishinari et al., 2024).

#### 445 *4.2. 3D food printing*

3D food printing is an advanced process combining 3D modeling, electromechanical 446 control, food science, and other technologies to create food products through an additive 447 manufacturing process (Wang et al., 2022). This method enables the production of customized 448 foods that cater to specific consumer needs, enhancing their sensory qualities, nutritive value, 449 and texture (Park et al., 2020). Foods such as chocolates, cakes, candies, and even artificial 450 meat have been successfully produced via 3D printing (He et al., 2020). Compared with 451 traditional food production methods, 3D printing offers benefits such as lower cost, simpler 452 operation, and faster production of customized products (Umeda et al., 2024). During printing, 453 gels undergo notable shear deformation because they are extruded through the printer nozzle. 454 The large deformation properties of gels, including their yield stress and viscoelasticity, impact 455 their ability to maintain and recover their shape after deposition for 3D printing. Gels that 456 cannot recover well may collapse or lose their structure, resulting in printed products of poor 457

quality. Therefore, gels need to be able to withstand large deformations while retaining their shape so as to ensure the successful 3D printing of complex food designs. Kim et al. (2022) developed a low-calorie form of surimi by combining carrageenan, a highly elastic protein substitute, with surimi through coaxial 3D printing. This process maintained the freshness and tenderness of surimi while reducing its caloric content, making it ideal for older adults or fitness-conscious individuals.

464 In recent years, 3D food printing has attracted significant attention, because of which natural food gels are being developed for printing. The deformability of these gels allows for 465 466 us to modify their fluidity, elasticity, and thermal properties through additives or treatments, enabling the fabrication of suitable materials for 3D printing. This technology can improve the 467 nutritional value of meals, benefiting both consumers without major health issues and those 468 with nutritional disorders such as malnutrition (Anukiruthika et al., 2020). Notably, 3D printers 469 can now create foods customized to consumer preferences by optimizing their flavor, texture, 470 color, price, and nutritional content. Natural substances, such as plant oils and emulsions, can 471 be used to modify the texture of printed food, influencing its printing behavior and gel 472 characteristics (Yang et al., 2024; Liang et al., 2024). Developing natural modifiers for 3D 473 printing can enable the development of customized food products that promote health and cater 474 to a wide range of consumers, from children to older adults (Das et al., 2023). 475

Gels require high elasticity and stress recovery to maintain structural integrity during extrusion. While shear-thinning additives provide improved printability, achieving consistent postdeposition stability remains challenging. Future research should focus on hybrid gels that offer both high printability and customized textural properties.

480 *4.3. Food preparation for individuals with dysphagia* 

481 Dysphagia is defined as the difficulty or inability to swallow liquids, solids, or semisolids,
 482 including drugs (Nishinari et al., 2023). This condition can lead to weight loss, malnutrition,

dehydration, and serious health issues such as aspiration pneumonia (Icht et al., 2018). 483 Individuals with dysphagia are advised to eat foods that are soft, moist, elastic, smooth, and 484 easy to swallow. Because gels are smooth, soft, and easy to break down, they are ideal for this 485 group of patients (Raheem et al., 2021). Gels prepared for patients with dysphagia should have 486 low fracture stress and should easily deform when chewed. Optimal large deformation 487 properties, such as high strain and low hardness, are important for controlling the texture and 488 489 consistency of these gels, making them easy to swallow without any risk of choking or aspiration. By adjusting the viscoelastic properties of food gels via additives, these gels can be 490 491 customized to meet the needs of patients with different levels of dysphagia, ensuring safe consumption. Thickeners modify the rheological properties of food, slowing its movement 492 through the mouth and throat, giving muscles more time to react and reducing the risk of 493 aspiration (Štreimikytė et al., 2020). Thus, gels can be used as thickeners to create softer, more 494 cohesive foods that are easier to chew and swallow. 495

Pure et al. (2021) modified the texture of a high-protein gel to make it suitable for patients with dysphagia. After microwave processing, the gel became a soft solid. Fei et al. (2024) developed a salmon-protein-based composite emulsion gel with konjac glucomannan and an emulsion filler. Konjac glucomannan altered the protein structure, and the filler improved the gel's elasticity and strength (Luo et al., 2024). These changes made the gel safer to swallow and improved its cohesion, enhanced its mouthfeel, and reduced its stickiness.

502 Soft gels with controllable deformation properties are essential for individuals with 503 swallowing difficulties. The current formulations focus on texture but often neglect sensory 504 aspects such as flavor release and bioavailability. Thus, studies integrating rheological testing 505 with sensory analysis are needed to optimize these products.

506 *4.4. Bionic foods* 

507

7 Gel-based bionic foods are an innovation in the food gel technology and can replicate the

structure and characteristics of natural foods (Zhou et al., 2023). They typically have a soft 508 texture and rich taste, and they mimic the flavor and texture of meats and other foods (Du et 509 510 al., 2023b). Plant-based meat alternatives are an important step in improving human health, conserving resources, and supporting animal welfare (Sun et al., 2021). These alternatives are 511 also rich in essential nutrients, making them a great choice for daily nutrition. Moreover, the 512 development of sustainable bionic foods, such as meat substitutes, is crucial to meeting the 513 514 growing dietary demands, particularly in light of resource scarcity and environmental concerns (Yang et al., 2023). Bionic foods can provide viable alternatives to natural foods, helping 515 516 mitigate these challenges (Du et al., 2022).

The ability of gels to deform without breaking is a key to simulating the texture of meat. 517 Bionic foods require gels that stretch, compress, and recover in ways similar to natural tissues. 518 Gel-based bionic foods that replicate the fibrous structure and chewiness of meat can be 519 developed by controlling the deformation properties, such as elasticity and toughness. This 520 replication is important for consumers' acceptance of plant-based alternatives. For example, 521 Du et al. (2023a) used agarose/konjac glucomannan double-network hydrogels to imitate the 522 texture of beef tripe. A 1:4 (w/w) ratio of agarose to konjac glucomannan resulted in a gel with 523 a texture similar to that of cooked beef reticulum and rumen, enabling the production of 524 artificial beef tripe. 525

526 Consumer surveys revealed widespread interest in the comparison of appearance, taste, 527 and texture of bionic foods with traditional ones (Hoek et al., 2011). While substantial progress 528 has been made in replicating the appearance and taste of natural foods by using flavor 529 compounds and plant pigments, the real challenge lies in recreating the texture, particularly 530 that of foods with complex structures. Through mechanical stretching, He et al. (2022) 531 developed an anisotropic hydrogel that successfully imitated the texture and taste of meat. 532 Likewise, for the simulation of the mouthfeel of other foods, bionic foods need to be processed to fully utilize the large deformation properties of gels. The replication of anisotropic textures
in meat analogs requires gels with directional deformation properties. While recent advances
in protein–polysaccharide composites show promise, scalability and environmental impact
remain critical challenges to solve.

537 *4.5. Food packaging* 

Hydrogels are useful as food packaging materials, particularly for controlling the moisture 538 539 content, which significantly impacts the shelf life of food products (Batista et al., 2019). These gels are referred to as colloidal gels because their molecules are evenly dispersed in water. 540 541 Their viscoelastic and structural properties are derived from polymer chains and their interactions with the surrounding medium (Ahmed et al., 2013). Packaging gels need to have 542 sufficient strength to withstand large deformations caused by handling, transport, and 543 environmental changes such as those in humidity. Their flexibility enables them to form a tight 544 seal around different food products, and their ability to retain water helps in moisture regulation, 545 extending the food's shelf life. 546

Hydrogels for packaging are made from natural polymers such as proteins, carbohydrates, 547 and lipids, which can be sourced from plants or animals (Du et al., 2021). The main natural 548 polymers used to prepare hydrogels for food packaging are presented in Table 2, along with 549 their key features and properties. The proportion of these natural polymers can be adjusted to 550 prepare hydrogels with the desired water retention capacity, flexibility, and mechanical 551 552 properties (Jildeh & Matouq, 2020). Notably, hydrogels with good malleability and swelling properties are suitable as food packing materials (Alharaty & Ramaswamy, 2020). Moreover, 553 these hydrogels need to exhibit good elasticity, tensile strength, and toughness, all of which 554 represent their large deformation properties. 555

556

## **Table 2.** Properties of natural polymer-based hydrogels used in food packaging applications

559 (Leyva-Jiménez et al., 2023).

Polymer	Material	Origin	Solubility	Advantages	Disadvantages
Polysaccharides	Chitosan	Chitin	Polar	Great mechanical	Highly hydrophilic
			solvents	properties; great	
				antimicrobial property;	
				gas/water barrier	
				properties	
	Starch	Plants	Water	Highly biocompatible	Highly hydrophilic,
				with polymers	brittle
	Pectin	Fruits	Water	High biodegradability;	Unstable mechanical
				biocompatibility with	properties
				polymers	
	Gum	Resins	Water	Emulsifying properties	Unstable mechanical
					properties; poor gas/water
					barrier properties
	Alginate	Marine	Water,	Self-healing properties;	Instability against
		algae	ethanol	formation of water	radiation or handling
				vapor barrier	
	Bacterial	Bacteria	Water	Film forming	Laborious process of
	cellulose			versatility; great	isolation
				gas/wate barrier	
Protein	Zein	Corn	Ethanol,	Film forming properties	Brittleness
			acetone		
	Soy protein	Soybean	Water,	Gas barrier properties;	Water resistance, rigidity,
	isolate		ethanol	pH- and temperature-	brittleness
				responsive films	
	Whey	Milk	Water,	Great mechanical	Moderate barrier to gas
			ethanol	properties; reduced	
				oxygen and carbon	
				dioxide permeability	

Continue

Polymer	Material	Origin	Solubility	Advantages	Disadvantages
Protein	Casein	Milk	Water	Forms flexible and	Brittleness
				nonopaque films; gas	
				and lipid barrier	
				properties	
	Wheat	Starch	Water	Moderate barrier to	Brittleness
				oxygen	
	Gelatin	Collagen	Water	Great gelling capacity;	Highly hydrophilic
				pH- and temperature-	
				responsive films	
Lipids	Wax	Bees	Water by	Low water vapor barrier	Poor mechanical
			emulsion		properties
	Acetoglycerides	Fatty	Water by	Stretch properties;	Low mechanical
		acid	emulsion	reduced water vapor	strength; low
				permeability	oxygen permeability
Another	Lignin	Wood	Water by	Antioxidant capacity;	Highly hydrophilic,
			emulsion	great mechanical	brittle
				properties	
	Poly lactic acid	Corn	Organic	Great mechanical	Brittle and weak
		biomass	solvents	properties	films

562 Despite their potential, the development of natural hydrogels for food packaging is still in 563 its nascent stages and several challenges remain to be addressed. This has limited their 564 widespread application in food packaging.

### 565 **5. Conclusions**

566 Gels are important in the field of food science. These materials have specific structural 567 and functional properties that enhance the texture, taste, and stability of associated food

products. A comprehensive understanding of the factors influencing the large deformation 568 properties of food gels, including gel concentration, pH, temperature, and additives, is crucial 569 to innovating and developing gel-based products. Theoretical models have undergone 570 continuous refinement and improvement ever since the discovery of the large deformation of 571 gels. This review highlights the different factors impacting the large deformation of gels and 572 also discusses the theories and mathematical models developed to understand and simulate this 573 574 process. Furthermore, it summarizes the application of the large deformation characteristics of gels in the fields of bionics and food science. 575

576 Although the application of gels with large deformation characteristics has grown in the food industry, several challenges continue to persist. The process of gel deformation is very 577 complex and is influenced by a multitude of factors. The deformation of food gels during the 578 process of eating cannot be fully simulated by mathematical models because these models 579 cannot capture the intricacies of the process. Food consumption does not only involve 580 mechanical actions such as chewing but also the enzymatic action of saliva and the electrical 581 signals that transmit sensory information to the brain. Currently, this multifaceted process 582 cannot be completely simulated through mathematical models or mechanical simulations. 583 These problems need to be addressed in future research. We believe future research should 584 focus on combining theoretical models with experimental data so as to more accurately predict 585 gel behavior and develop blended gels customized for specific applications, such as 3D printing 586 and the fabrication of dysphagia-friendly foods. Sustainable and scalable innovations, such as 587 bio-based crosslinkers and mixed additive systems, should also be explored. 588

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#### 594 **CRediT authorship contribution statement**

- 595 Bing Hu: Conceptualization, Formal analysis, Investigation, Writing–review and editing
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- 597 Lingyu Han: Conceptualization, Methodology, Writing-review and editing
- 598 Yiguo Zhao: Formal analysis, Writing–review and editing
- 599 Cunzhi Zhang: Methodology
- 600 Jijuan Cao: Conceptualization, Formal analysis, Writing-review and editing
- 601 Jinxin Yang: Methodology, Software
- 602 Yapeng Fang: Methodology
- 603 Declaration of competing interest
- 604 The authors declare that they have no conflicts of interest regarding this study.

### 606 Figure captions

- 607 **Fig. 1.** Rheological responses in viscoelastic solids.
- 608 Fig. 2. Hydrogel undergoing deformation in response to external stimuli, and changes from the
- 609 reference state  $X_K$  to the current state  $x_i$ .
- 610 Note: Image created using BioRender and published with permission
- 611 **Fig. 3.** Applications of food gels.
- Note: Image created using BioRender and published with permission

613







Fig. 3

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