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Current Opinion in
Food Science

# Gels: model systems for soft matter food physics

Thomas A Vilgis

This review summarizes recent work on edible (food) gels. The main goal is to relate the structural properties of different gels and textural properties in physical terms. Therefore agarose is used in combinations with other non-gelling hydrocolloids and low molecular carbohydrates to find basic relations between the long and short range interactions in food systems, which offers insight the interplay between gelling and non-gelling hydrocolloids. As non-gelling agents xanthan and alginate are used, since they provide significant differences in their chain stiffness and charge distribution along their polymeric backbone. Both show different impacts on the low and high shear rate modulus, which are related to the textural properties of the gels. The impact of low molecular weight sugars, sucrose and trehalose is also taken into account. The local hydrogen bond interactions together with the position and orientation of the hydroxyl-groups of the sugars influence the macroscopic moduli in a systematic way. The review covers therefore the influence of local and mesoscale interactions on the elastic properties on food gels.

**Address**
Max-Planck-Institute for Polymer Research, Ackermannweg 10, 55129 Mainz, Germany

**Current Opinion in Food Science** 2015, **3**:71–84

This review comes from a themed issue on **Food physics and material science**

Edited by **Ruud van der Sman**

For a complete overview see the Issue and the Editorial

Available online 31st May 2015

**http://dx.doi.org/10.1016/j.cofs.2015.05.009**

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## Introduction

Soft gels can be considered as simple models for foods, which enable, due to the low number of components, a systematic study and understanding on the short and long range interactions, the influence of chain flexibility, and molecular weight, which are in real foods not easily accessible. The physics of viscosity enhancement (thickening) and gelation in food systems is another fundamental problem, since it relates molecular structure and food texture on the basis of model systems, which allow more systematic studies. Simple water based gels can be considered as physical model systems for foods. Their number of components, mainly water, gelling agents, eventually sugars or salts and aroma compounds is limited. There exists a large variety of hydrocolloids from plant cells, such as agarose, alginate, carrageenan, pectin, or microbiological fermented (starch) products, as gellan and xanthan, or cellulose derivatives [1•,2]. Their differences in molecular structure and molecular weight provide a sound basis for structuring liquids and gels in physically more systematic way compared to natural foods. The selection of such gelling agents with their different properties allow to relate molecular properties, which define the structure of the gels to their function and mouth-feel of the macroscopic fluid or gel. For example, gelatin from the collagen of different animals can be considered as an helix forming amphiphilic polymer [3••], which appear soft elastic, gellan is a charged gelling agent, which forms stronger, but more brittle gels [4,5]. Alginate and pectin can be considered as partially charged triblock copolymers which form with bivalent ions gels via the so called egg box model [6]. Their gel properties can be controlled by the alginate structure, for example, the block length and the choice of the bivalent ions [7]. The thickening properties of stiff macromolecules like xanthan are very different compared to those of guar gum, alginate, or carrageenan, due to differences in charged groups, polar repeat units or chain flexibility [1•,8••].

Hydrocolloids are in general water-soluble polymers [9–11], which change the rheological and textural behavior of water based fluids according to their molecular properties. In food systems hydrocolloids are used for enhancing the textural properties. For example, creams with specially controlled flow properties stay longer in the mouth and release their taste slower but longer, so that typical flow times for certain molecular processes match perception relevant time scales in the mouth. Such physical parameters are relevant for sensory science. In many cases the use of hydrocolloids are used to stabilize the mouth-feeling of a certain product under the natural and possible variation of physical parameters, such as acidity, salt or sugar content and basically the temperature, under which the product is eaten, for example, directly out of the fridge or after some warming at room temperature. The taste receptors those for sour, sweet, bitter, salty, umami and fatty taste are for quite some time longer under the influence of the molecules which trigger them, the taste ecomes enhanced. A more secondary effect is more important. During the time the food stays in the mouth, the temperature of the liquid changes. During this process more of the volatile compounds responsible for the retronasal effects [12] may escape from the liquid and evaporate. Hence the sensory effects are also enhanced.

These naive thoughts suggest a number of physical requirements for a simple theory for the structure property taste relationship: Firstly, the description of the rheological flow properties, via the viscosity and the physical behavior of the fluid and finally, a more detailed description of possible mechanisms of the taste release. However, since their use in 'molecular cooking' their special features became available to new applications and the products are used on a practically basis by a broader public [13].

The elementary approaches of viscosity enhancement by adding colloids goes even back to Einstein's [14],

$$\eta = \eta_0(1 + \frac{5}{2}\phi + \ldots) \qquad (1)$$

where $\eta_0$ is the viscosity of the solvent and $\phi$ is the volume fraction of the added spherical colloids dissolved. The shape of the colloids determines only numerical factors, and for strong dilution hydrocolloid polymers with a polymerization degree $N$ and a hydrodynamic radius $R \propto bN^{\nu}$, and a volume fraction $\phi = n/R^3$, $n$ being the number density of the chains as well, as long as the chains do not overlap [15••]. When the hydrocolloid with a chain length $N$ are sufficiently flexible, for example, polar, such as non-ionic hydrocolloids (e.g. methyl or carboxyl methyl celluloses), well-know results from polymer physics (see, e.g. [15••]), such as the Rouse scaling, $\eta \propto k_B TN$ can be adopted. Above the overlap concentration yield a stronger rise of the viscosity, which lead to chain length dependence $\eta \propto N^3$.

In many other, more recent, phenomenological approaches, empirical equations have been proposed to describe the increase of viscosity with concentrations, molecular weight or structure of the dissolved polymers, which can be found, for example, in [16•]. Amongst them the Mark–Houwink relation should be mentioned, which finds many applications in food hydrocolloids, see, for example, [17,18] for some recent references.

Charged chains can be theoretically described by polyelectrolyte chains [19••,20,21•]. The polyelectrolyte chains appear in salt free solutions sufficiently stretched, depending on their charge. Simple Flory type scaling suggests therefore [19••,21•] for salt free solutions

$$R \simeq b\left(\frac{l_B f^2}{b}\right)^{1/3} N \qquad (2)$$

where $b$ is the mean size of the repeat unit, $l_B$ is the Bjerrum length and $f$ the charge fraction, which is the number of charged monomers in the chain. Thus the chain extension is proportional to the chain length itself, but on scales below the 'blob size' $\xi = b(b/l_B f^2)^{1/3}$ the conformation is coiled. Ionic (hydrocolloid) chains become more 'stiffer' when immersed in the food preparations, which yields strong impact the mouth feel.

In this paper some recent work is reviewed. The main emphasis is put on the combination of gel forming (here agarose), non-gel forming hydrocolloids, such as xanthan, a relatively stiff polyelectrolyte and alginate, a relative flexible block polyelectrolyte, and low molecular weight sugars (sucrose and trehalose) as simple models systems for food-like soft matter. The choice has been made deliberately: The gel forming agarose ensures the solidification above the gel point concentration, whereas the non-gelling hydrocolloids act as mobile phase in the networks, which are able to change the rheological behavior in a different way. Both are polyelectrolyte molecules, but with significantly different stiffness. In order to act the alginate as simple thickener, bivalent ions (salts) need to be avoided, otherwise alginate forms a gel as well (see, e.g. Ref. [22]. In the next paragraphs below a model for the special thickening of the rod-like molecules xanthan is discusses, before coming to gels and composed gels. The difference between thickening and gelling can be discussed at the viscosity and the shear modulus. Thickeners increase the viscosity whereas gelformers show (in general) first an increase of the viscosity when the gel point is approached, which becomes (theoretically) infinite at the gel point and shows a finite elastic modulus above the gel point. The last sections are devoted to low molecular weight carbohydrates (sugars), where it is shown on experimental grounds that the orientation and location of the hydroxyl groups provide different nature of hydration shells. These local differences influence the macroscopic properties significantly.

## Salts, thickening and chains conformation

As well known from flexible and semi-flexible polyelectrolytes, salts, and the increase of the ionic strength has severe impact on the chains conformation and thus the thickening. In the presence of salt interactions between charges become screened and the Coulomb repulsion becomes a Debye–Hückel screened potential between two charges

$$U_C(\mathbf{r}) \propto \frac{1}{r}\mathbf{xrarr;U_{DH}}(\mathbf{r}) \propto \frac{\exp(-\kappa\mathbf{r})}{\mathbf{r}} \qquad (3)$$

where $\kappa^2 = 4\pi l_B I$, $I$ being the total ionic strength, $r$ denotes the distance between two charges. The electrostatic screening length $\xi = 1/\kappa$. The screening effect has indeed some relevance for culinary applications of any thickening (or gelling) agent. Since the presence of salt in any liquid like sauces or broth will screen the interaction, it will change the shape of the polymer and thus its effect on viscosity. The size of the chains in a higher salted solution shrinks [23,24•]. It was shown that the persistence length $l_p$ of the chain in salted solution is of the order of the electrostatic blob size, that is, $l_p \simeq 1/\kappa$ and scales this with $l_p \propto 1/I^{1/2}$. The persistence length decreases with increasing ionic strength. All interactions are screened at very high salt concentrations, the interaction potential from Equation (3) becomes short ranged.

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