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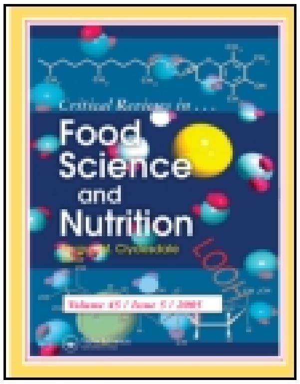

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Food Gels: Gelling Process and New Applications

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Food Gels: Gelling Process and New Applications

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Food gels are viscoelastic substances and several gelled products are manufactured throughout the world. The gelling agents in foods are usually polysaccharides and proteins. In food gels, the polymer molecules are not cross-linked by covalent bonds with the exception of disulphide bonds in some protein gels. Instead, the molecules are held together by a combination of weak inter-molecular forces like hydrogen bonds, electrostatic forces, Van der Waals forces, and hydrophobic interactions. Polysaccharides including hydrocolloids are strongly hydrated in aqueous medium but they tend to have less ordered structures. The mechanism of gelation depends on the nature of the gelling agent(s) and on the conditions of gel formation like the temperature, the presence of ions, the pH, and the concentration of gelling agents, etc. Characterization of gels can be performed in several ways of which rheological measurements are frequently practiced. Multi-component or mixed gel system is an important area of interest in which two or more gelling components are simultaneously used to achieve certain specific structural and functional characteristics. We here discuss about the different gels and gelling agents, the characterization of gels, and the mechanism of gelation with an emphasis on mixed or multi-component gels that would have significant commercial applications.

Keywords Gel, gelling agents, multi-component gels, mechanism, hydrocolloids

INTRODUCTION

A gel is an intermediate between a solid and liquid possessing both elastic (solid) and flow (liquid) characteristics. A sol (liquid) to gel (solid) transition takes place when gels are formed. Several gel forming ingredients are selectively employed to generate food gels of acceptable attributes, particularly the extent of textural properties, which is an indication of the process of gelling.

A number of foods are marketed in the form of gel that offers convenience to the consumers. Examples include jam, jelly, confectionery products, desserts, quick-set gels, and products based on fruits and vegetables. Invariably, one or more gelling agents are employed to achieve the desired or targeted parameter(s). The gelling agents that are frequently used in such products are usually permitted food hydrocolloids. These hydrocolloids are derived from natural sources and refers to a range of polysaccharides and proteins that are nowadays widely used in a variety of industrial sectors to perform a number of functions including thickening and gelling aqueous dispersions, stabilizing foams, emulsions, and dispersions, suspend particulate materials and inhibit or decrease syneresis, as well as increase water retention (Sutherland, 2007). In addition, polysaccharides are incorporated because of their capacity to control the texture of foods and for the controlled release of flavors; the advantage is also taken from the ability of some mixtures of polysaccharides to exhibit synergistic gelling. Associated with these readily measurable physical properties are others such as mouthfeel or sensory characteristics, which also show some correlation with physical properties.

A food gel can also be considered as a high moisture three-dimensional polymeric network that resists flow under pressure and more or less retains their distinct structural shape (mechanical rigidity). It is a viscoelastic system with a storage modulus (G') larger than the loss modulus (G''). It is a continuous network of interconnected particles or assorted macromolecules dispersed in a continuous liquid phase. Gellification is the phenomenon involving the association or cross-linking of the polymer chains to form a three-dimensional network that traps or immobilizes water within it to form a rigid structure.

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GELLING AGENTS

Gelling agents are food additives used to thicken and stabilize various foods, like jellies, desserts, and candies. These agents provide the foods with texture through formation of a gel. Some stabilizers and thickening agents are also gelling agents.

Only a comparatively few gums form gels, and these vary widely in gel character and texture so that they are used for specific food applications. Frequently used gelling agents include natural gums, proteins, starches, pectins, and agar-agar, and can be broadly classified into polysaccharides and proteins. Most gelling polysaccharides are hetero-polysaccharides and hydrocolloid in nature (Table 1). They can be irregular unbranched structures, regular or irregular branched structures block copolymers, or structures containing complex but well-defined repeating units. Their sources are also different including plant, animal, and microbial origin. Though possibly all of them can be used for the purpose of thickening, the emphasis is on gel formation. The applications include jams, jellies, salad dressings, deserts, marmalade, jujubes, yogurts, etc. On the other hand, several proteins are also used for the purpose of gel making (Table 2). These include zein from corn, and several animal proteins like gelatin and whey proteins.

CONDITIONS OF GEL FORMATION

The gel formation is a spontaneous process from a simple polymer dispersion, or particulate suspension, and externally controllable conditions of temperature or solution composition. Thus, the sol-gel conversion process usually involves aggregation of particles, or macromolecules, with eventual generation of a network spanning the entire container volume (Clark, 1992).

The formation of gels can be classified broadly as physically induced (heat, pressure) and chemically induced (acid, ionic, enzymatic) gelation reactions. In case of protein gels, the gelation of proteins requires a driving force to unfold the native protein structure, followed by an aggregation process giving a three-dimensional organized network of aggregates or strands of molecules cross-linked by non-covalent bonds or less frequently by covalent bonds (Totosaus et al., 2002). The conditions of gel formation depend mostly on the several physico-chemical factors as mentioned in the subsequent sections.

Temperature

Heat-induced gelation is probably the most important and common method to obtain gels (Aguilera and Rademacher, 2004). Gelation is a two-step process; an unfolding or dissociation of the molecules due to the energy input takes place in the beginning to expose reactive sites. The second step is the association and aggregation of unfolded molecules to form complexes of higher molecular weight. The first step may be reversible while the second one is usually an irreversible process. Presumably, disulphide (-S-S-) bridges and hydrophobic interactions often play major roles. The overall reaction rate can be determined either by the unfolding or by the aggregation reaction depending on the ratio of the reaction rates of the single steps in a particular temperature range.

Pressure

High pressure offers an additional degree of freedom in modifying functional properties of molecules since high pressure can be applied as a single process or in combination with others, in particular with increased temperatures. In general, high pressure favors reactions, which lead to a reduction of the overall volume of the system. Pressure causes water to dissociate and the pH becomes more acidic under pressure. Differences exists in appearance and rheological properties of gels made with the application of pressure and heat (Heremans, 1995).

Ionic Strength

Monovalent and divalent cations such as sodium and calcium can increase the ionic strength of the gel. The electrostatic repulsive forces between the molecules are reduced or neutralised and gelation can occur. Ionic-induced gelation has been reported for pre-denatured whey proteins and in contrast to heat-induced gelation, is named cold gelation (Bryant and McClements, 1998; Alting et al., 2002). Ionic-induced gelation is of greater importance in polysaccharide gels, for example, alginate, pectin, or carrageenan.

pH

Changes in pH due to the addition of acids or microbial fermentation change the net charge of the molecule and therefore alter the attractive and repulsive forces between molecules as well as the interactions between molecules and solvent, that is, hydration properties. In addition, the solubility of salts changes with pH which may contribute to gel formation. The mechanism of acid gel formation could be explained by the fractal aggregation theory (Lucey and Singh, 1997).

Presence of Enzyme

Enzyme-induced gelation is based on the introduction of artificial covalent cross-links into food proteins. Among others, reactions catalyzed by trans-glutaminase (TG), peroxidase, and polyphenol oxidase are suitable for cross-linking of proteins (Lauber et al., 2003).

Solvent Quality

The nature and presence of solvent markedly influences gel formation, for example, the concentrated sugar solution is a

Table 1 Hydrocolloids used as gelling agents

Gelling agent	Source	Name of binding blocks	Applications	References
Agar	Red algae (*Gelidium* sp. and *Gracilaria* sp.) or seaweeds (*Sphaerococcus euchema*)	Agaropectin and agarose (D-galactose and L-anhydrogalactose)	Used as laxative, vegetarian gelatin substitute, in jellies and Japanese desserts such as anmitsu	Armisen et al., (2000); Stanley (2006)
Cereal flour and starch (cooked/ instant/pre-gelatinized/ modified)	Potato, wheat, rice, maize, tapioca	D-α, 1–4, 1–6 linked glucose polymer, mainly made of amylose (linear) and amylopectin (branched)	Secondary gelling agent, cost effective, rice flour based gels	Jena and Bhattacharya (2003); Alka and Bhattacharya (2008)
Cellulose (common forms are carboxymethyl-cellulose)	Plant cell wall material and chemically modified	Homo-polymer of β (1, 4) D-glucose	Salad dressings and deserts	Williams and Philips (2000)
Carrageenan (kappa, iota, lamda, hybrid, blend, refined)	Red seaweeds (*Chondrus crispus*)	Sulphated D-galactose and L-anhydrogalactose	Desserts, gel to immobilise cells/ enzymes	Williams and Philips (2000)
Pectin (high-methoxyl, HM and low-methoxyl, LM)	Hetero polysaccharide derived from the cell wall of higher terrestrial plants, and fruits like citrus peel, guava, apple	Linear polymer of partly esterified α-(1–4)-linked D-galactouronic and mannuronic acid. Sometime galacturonic acid is replaced by rhamnose, galactose and arabinose	Jam, jelly, marmalade, jujubes, yogurt	Williams and Philips (2000); Oakenfull (1987)
Guar gum	Endosperm of guar gum	Linear chain of Galactomannan unit	Pastry fillings, yogurt, liquid cheese products and sweet desserts	Cui et al. (2007); Richardson et al. (1998)
Gum Arabic	Sap taken from two species of the *Acacia* tree, *Acacia senegal* and *Acacia seyal*	Complex mixture of saccharides and glycoproteins	Hard gummy candies, chocolate candies and chewing gums	Sutherland (2007)
Xanthan gum	Fermentation of glucose or sucrose by *Xanthomonas campestris*	Polysaccharide chain consists of two β-D-glucose units linked through the 1, 4 positions. The side chain consists of two mannose and one glucouronic acid, and thus the chain consists of repeating modules of five sugar units	Salad dressings and sauces, helps to stabilize the colloidal oil and solid components against creaming by acting as an emulsifier and texture modifiers in different foods	Sutherland (2007); Doublier and Cuvelier (2006)
Locust bean gum/Carob gum	Extracted from the seeds of the Carob tree	It is a galactomannan	Gelling agent	Cui et al. (2007); Schorsch et al. (1997)
Alginate (alginic acid)	Brown seaweeds (*Macrocystis pyrifera, Ascophyllum nodosum* and various types of *Laminaria*)	Linear copolymer with blocks of (1–4)-linked β-D-mannuronate and its C-5 epimer α-L-guluronate residues, covalently linked together in different sequences	Jellies, gelation with divalent cations, cell immobilization and encapsulation, appetite suppressant	Roopa and Bhattacharya (2008)
Konjac mannan	Tubers of Konjac (*Lasioideae amorphophallus*)	Glucomannan is heteropolysaccharide consisting of D-glucose (G) and D-mannose (M), linked by β -D-1,4 bonds with a G/M ratio of 1 to 1.6	Gelling, texturing, and water binding agent, to provide fat replacement properties in fat-free and low-fat meat products	Takigami (2000)
Gum Karaya	Extract of S*terculia* trees	Acetylated acidic polysaccharide containing α-D-galacturonic acid and α-L-rhamnose residues as the main chains and the acid is linked by 1, 2 linkage of β-D-galactose or by 1, 3 linkage of β-D-glucuronic acid on side chains	Brown sauce, toppings, fillings	Le Cerf et al. (1990); Weiping and Branwell (2000)
Gum Tragacanth	Dried sap of several species of legumes of the genus *Astragalus,* including A. *adscendens,* A. *gummifer*, and A. *tragacanthus*	Consists of two fractions: a water-soluble, neutral arabinogalactan called tragacanthin and an insoluble fraction called tragacanthic acid	Confectionary icings, dressings	Aspinall and Bail Lie (1963)

poor solvent for pectin. Hydrogen bonds in the junction zones can only be formed in concentrated sugar solution, and hence, gelation takes place only in this type of solvent (Clark, 1992; Richardson et al., 1998).

Concentration of Gelling Agent

Gel formation only occurs above a critical minimum concentration, C^*, which is specific for each hydrocolloid. Agarose

Table 2 Proteins used as gelling agents

Gelling agent	Source	Name of binding blocks	Applications	References
Gelatin (acidic/alkaline)	Animal skin and bones (made by partial hydrolysis of collagen of animal connective tissue)	Protein high in glycine and proline	Gelling agent in gelatin desserts, jelly, trifles and confectioneries, jam, yogurt, cream cheese and margarine	Ward and Courts (1977); Schrieber and Gareis (2007); Oakenfull (1987)
Whey protein	Acid or sweet dairy whey, separated from the casein curd as the soluble fraction during cheese manufacture	Mainly comprise of globular proteins like β-lactoglobulin and α-lactalbumin	Gelling agent and thickener in food industry	Aguilera and Rademacher (2004)
Soya proteins	Soybeans	Interaction of two globulin proteins like glycinin and β-conglycinin	Heat set gel	Bhattacharya and Jena (2007)
Egg proteins	Egg	Consists of about 70% albumen (globular proteins containing ovomucin fibres) and 30% egg yolk (several types of low-density lipoproteins)	Gelling and thickening agent for confectionery products	Aguilera and Rademacher (2004)
Zein	Corn	Peptide chain containing mainly prolamine	Gel coated candy, nuts, fruit, pills and other encapsulated food and bakery products	Shahidi and Synowiecki (1991)

will form gels at concentration as low as 0.2% while for acid thinned starch, a concentration of ∼15% is required before gels are formed (Aguilera and Rademacher, 2004).

Molar Mass/Degree of Polymerization

If concentration of the polymer is well above the critical concentration, the effect of molar mass is insignificant, but if the concentration is in the range of C* values, the higher the molar mass, the higher is the modulus and faster is the process of gelation (Walstra, 2003).

CHARACTERIZATION OF GEL

Gel formation is basically a transformation of sol to the state of gel during which the viscoelasticity changes abruptly with simultaneous development of solid characteristics. In other words, there is an interchange of continuous and discontinuous phases during gel formation. Thus, there is a necessity for the measurement of viscoelastic (rheological) characteristics of material which is mostly conducted by different type of experiments (Table 3). On the other hand, Table 4 shows the non-rheological methods for the measurement of gel characteristics. These are generation of heat flow data and microscopic observations.

Table 3 Methods for the rheological measurement of gel characteristics

Nature of test	Type of measurement	Instrument used	Measurement parameters	Applications	References
Fundamental tests	Compression	Texture measuring system	Modulus of elasticity, Poission's ratio	Surimi gel	Kim et al. (2006); Macdonald and Hamman (1992); Walstra (2003)
	Stress relaxation	Texture measuring system	Residual stress, relaxation time	Gellan gels	Morris (1986)
	Creep	Controlled stress rheometer	Shear modulus, creep compliance	Soy and gelatin gels	Kamata and Kinsella (1989); Chronakis et al. (1995)
	Oscillation	Controlled stress rheometer	Storage modulus (G′), loss modulus (G″), phase angle, complex modulus and viscosity	Viscoelastic characterization of rice, soy gels, mixed gels	Jena and Bhattacharya (2003); Kim et al. (2006); Bhattacharya and Jena (2007); Keogh et al. (1995)
Empirical	Puncture force	Texture measuring system	Puncture characteristics	Characterization of rice gel	Jena and Bhattacharya (2003); Kim et al. (2006)
	Compression	Texture measuring system	Peak force, firmness, compression energy	Measurement of gel quality and gel strength	Kim et al. (2006); Smewing (1999)
Imitative	Texture profile analysis (TPA)	Texture measuring system	Parameters of texture profile analysis like hardness, brittleness, adhesiveness, springiness, cohesiveness	Food gels	Pons and Fiszman (1996)

Table 4 Other methods for the measurement of gel characteristics

Type of measurement	Instrument used	Measurement parameters	Applications	References
Structural Characterization	Differential scanning calorimeter (DSC)	Heat flow	Gellan and polyvinyl alcohol blend film	Sudhamani et al. (2003)
	X-ray diffraction and SAXS	Particle size analysis	Nano delivery system in food	Luykx et al., (2008)
	Colorimeter	Color measurement	Gellan edible films	Leon et al. (2008)
Microscopic characterization	Light microscopy (LM)	Area of the granules	Tapioca starch gel	Vittadini et al. (2006)
	Scanning electron microscope (SEM)	Structural arrangement of components	The number, area and location of particles in gel	Moritaka et al. (2003)
	Transmission electron microscopy (TEM)	Structural distribution of constituents	Characteristic studies of mixed gel	Aguilera and Stanley (1999)
	Atomic force microscope	Structure of the molecules	Structural characteristics of nanoparticles	Luykx et al. (2008)
Molecular Characterization	NMR	Conformation changes on gelation	Structural features of the constituents	Saito (1995)
	FTIR	Molecular structure	Infrared spectra of the components	Sudhamani et al. (2003)
	FT-RAMAN; Near infrared resonance	Molecular characterization	Functional characteristics of pectins	Wilats et al. (2006)
Proximate analysis	Vacuum oven	Moisture	Moisture estimation	Leon et al. (2008)
	Atomic absorption spectroscopy	Mineral content	Chitosan and whey protein isolate based model system	Laplante et al.. (2005)
	Kjeldhal apparatus	Protein	Whey protein–cassava starch gel	Aguilera and Stanley (1999)
	DNS method	Carbohydrate	Sugar content of the gel	Aguilera and Baffico (1997)

Fundamental tests (Table 3) do not depend on the geometry of the sample and the instrument used. The rheological properties determined at large deformations include fracture, failure, and rupture characteristics like stress/strain, and are usually determined by uniaxial compression and tension. Rheological or textural characteristics of starch gels have been investigated mostly on model systems in order to understand basic factors and mechanism involved in the gelling and characterize the gel properties (Doublier et al., 1992; Jena and Bhattacharya, 2003).

The Rayleigh scattering measurement can be employed to estimate the size of the particle in solution. It can provide information on the various aggregation processes occurring during gelation. Besides, dynamic light scattering can also be used to study the rheology of gelatine dispersion by the measurement of the diffusion coefficient of suspended particles and the effective viscosity of the gel (Vittadini et al., 2006).

Rheological properties depend on the presence of molecular network. Measurement can be made which shows the relationship between stress (force per unit area) and strain (deformation due to applied force) for a gel under compression. The Young's or elastic modulus is the ratio of stress to strain of a material when tested within the linear limit of elasticity. The maximum stress that the gel can sustain is rupture strength (RS). Bulk modulus (K) can be obtained when the force is applied from all the directions (isotropically) and the change in volume per original volume is obtained (Kim et al., 2006).

Rheological measurement of gel characteristics has been broadly categorized into small and large deformation tests (Van Vliet, 1995). Small deformation test generally implies structural features while large deformation tests are employed to measure the stress, strain, and failure properties of a gel (Ross-Murphy, 1995).

TYPES OF GEL

The polymer molecules in gels are not cross linked by covalent bonds (except protein gels); instead, the molecules are held together by weak intermolecular force such as hydrogen bonds, electrostatic forces, Van der Waals forces, and hydrophobic interactions. The gelation process depends on the interaction of two or more polymer molecules forming gel (Aguilera and Rademacher, 2004). There are different kinds of gel depending on their structural features and characteristics.

Hydrogel

It is a network of polymer chains that are water-insoluble, sometimes found as a colloidal gel in which water is the dispersion medium. They are super absorbent (contain over 99% water) natural or synthetic polymers. Hydrogels possess also a degree of flexibility very similar to natural tissue due to their high water content. Environmentally sensitive hydrogels have the ability to sense changes of pH, temperature, or the concentration of metabolite and release their load as a result of such a change (Kong, 2005). Natural hydrogel materials include agarose, methyl cellulose, and other naturally derived polymers. These hydrogels are used in sustained release delivery systems for drugs and nutrients.

Organogels

It is a non-crystalline, non-glassy, thermoreversible solid material composed of a liquid organic phase entrapped in a structuring network (Terech, 1997). The liquid can be an organic solvent, a mineral oil, or a vegetable oil. The solubility and particle dimensions of the structurant are important characteristics for the elastic properties, firmness, and integrity (Van Esch et al., 1999). An example of formation of an undesired thermoreversible network is the occurrence of wax crystallization in crude oil (Kumar and Katare, 2005).

Xerogels

A xerogel is a solid formed from a gel by drying with unhindered shrinkage. Xerogels usually retain high porosity (25%) and enormous surface area (150–900 m^2/g), along with very small pore size (1–10 nm) (Walter, 1998).

Aerogels

It is a colloidal gel in which gas is used as the dispersion medium (Zúñiga and Aguilera, 2008). When solvent removal occurs under hypercritical (supercritical) conditions, the network does not shrink and a highly porous, low-density material known as an aerogel is produced. These materials mostly possess exceptional properties including very low density, high specific surface areas and excellent thermal insulation properties.

Weak Gels

Some polysaccharides, often of food origin, show properties intermediate between polysaccharide solutions and true gels, and form weak gels. Under low deformation, weak gels behave as elastic gels. At sufficiently large deformation or at high shear rates, they fracture, break irreversibly, and flow. There are a number of polysaccharides such as xanthan gum and gellan gum that exhibit this type of behavior (Mohammed et al., 2007).

Fluid Gels

Gels occur when hot hydrocolloids dispersions are allowed to cool and set under quiescent conditions (Valli and Miskiel, 2001). The dispersion separates into polymer-rich micro-particles and polymer-poor regions, which forms the interstitial space between the particles. The overall rheology of the system comes from particle-particle interactions, which can be disrupted by low shear rates and shear stresses (Norton et al., 1999; 2006). These systems, commonly known as fluid gels, can have a wide range of textures including a light pourable gel and a thicker spreadable paste. Rheologically, the low-shear viscosity of gellan gum gels is relatively high at a low gum concentration and thus the formation of fluid gel is possible. Polysaccharides like agar and k-carrageenan form gels under suitable shear field during gelation along with other liquid foods (Norton et al., 1999).

Temperature Sensitive Gel

Gels can also be thermoreversible or thermoirreversible. Thermoreversible gels are those which melt on heating and gel upon cooling (e.g., agar, carrageenan, gellan, and gelatin) while thermo-irreversible gels are those which once formed on heating will not melt (e.g., alginate, low methoxyl pectin). Based on temperature dependence of the elastic modulus, polysaccharide gels may be classified into four categories (Nishinari and Zhang, 2004). These are (a) cold set gels like agarose, carrageenan, and gellan which form a gel on cooling the dispersion (b) heat set gels like some cellulose derivatives (MC and HPMC), curdlan, konjac glucomannan which form gel on heating the dispersion (c) re-entrant gels like xyloglucan which forms gels at intermediate specific temperature ranges and remain in the sol state at outside of this temperature range, and (d) inverse re-entrant gels like a mixed solution of methyl cellulose and gelatin which forms gel at higher and lower temperature, and stays in the sol state in the intermediate temperature range.

MECHANISM OF GEL FORMATION

It is desirable to understand the mechanism of gelling processes in order to achieve gels of specific attributes. The mechanism of gelation mainly depends on the gelling agents, nature, and synergistic effects of different constituents.

Gelatin

Gelatin melts when heated and solidifies when cooled again. Together with water, it forms a semi-solid colloidal gel. Gelation is governed by the partial reformation of triple helices found in collagen during cooling. In the first step, a polypeptide chain takes an orientation to induce a reactive site. Later, condensation of two other chains near the reactive site occur giving rise to triple helix formation (Renard et al., 2006).

Whey Proteins

Heat-induced gelation of whey proteins is typical of globular proteins and proceeds through a series of transitions, such as (i) denaturation (unfolding) of native proteins, (ii) aggregation of unfolded molecules, (iii) strand formation from aggregates, and (iv) association of strands into a network. Aggregates are formed in the presence of salts (Cayot and Lorient, 1997; Jong et al., 2009).

Soy Proteins

Gelation is achieved by heating soybean flour (Bhattacharya and Jena, 2007) or milk followed by addition of salt (e.g., Ca^{++} or Mg^{++}) to form a gel or curd, although acidification (e.g., with glucono-δ-lactone) also induces aggregation of denatured protein molecules.

Milk Proteins

Casein molecules are strongly hydrophobic, and thus submicelles are held together by hydrophobic bonds and salt bridges. Enzymatic hydrolysis of k-casein by rennet releases CMP (caseinomacropeptide) and causes the micelles to aggregate leading to rennet gelation (Keogh et al., 1996; Fertsch et al., 2003).

Egg Proteins

Both albumen and yolk of liquid eggs have the capacity to form gels upon heating. Gel formation is a two-step process of denaturation followed by aggregation of denatured proteins (Montejano et al., 1984; Woodward and Cotterill, 1986).

Alginates

Gels are formed on the addition of polyvalent cations at a low pH (< 4). Guluronic acid residues give a buckled conformation providing an effective binding site for the cations (Picullel et al., 1994). The gel strength also depends on the nature of the divalent cation with the order like $Ba^{++} > Sr^{++} > Ca^{++} > Mg^{++}$ (Smidsrod, 1974). In contrast to most gelling polysaccharides, alginate gels have the particular feature of being cold setting.

Pectin

The gelling characteristics of pectin strongly depend on the degree of esterification. High methoxyl pectins will gel only in the presence of sugars or other co-solutes (such as sugars, polyols, or monohydric alcohols) (Oakenfull and Scott, 1984), and at a sufficiently low pH (3.0–4.5). Gelation of low mehoxyl pectins occurs only in the presence of divalent cations like calcium.

Agar

Gelation of agar is a thermo-reversible process and takes place only due to the formation of hydrogen bond (Armisen et al., 2000). Agar requires heat to bring it into dispersion. On cooling, the hot dispersions set to gels. Molecules undergo a coil helix transition followed by aggregation of helices (Stanley, 2006).

Starch

When native starch grains are heated above a certain critical temperature (gelatinization temperature) in an excess of water, gelatinization occurs, which implies that amylose leaches from the granules and that the latter greatly swell and eventually fall apart. Irreversible swelling of the granules then occurs accompanied by solubilization of amylose. If the system then is cooled and the amylose concentration is above the chain overlap concentration c*, corresponding to 2–4% starch (depending on starch type), a gel is formed. This is mainly due to the formation of microcrystallites of amylose and the gel resembles a polymer gel. Upon cooling, a gel is formed, consisting of closely packed granules, with a thin layer of amylose gel as a kind of glue between the granules (Murphy, 2000; Sikora et al., 2007; Viturawong et al., 2008).

Carragennan

Carrageenan is an ionic polymer and forms helical gels on cooling in presence of salts (electrolytes) particularly K^+ ions. Certain cations like K^+, Rb^+, Cs^+, and $NH_4{}^+$ promote both helix formation and gelation. Molecules undergo a coil helix transition followed by aggregation of helices (Wang et al., 2005). The gelation of carrageenan generally involves association of the polymer chains by the formation of intermolecular double helices to form "ordered domains." Gelation occurs with the subsequent aggregation of these domains mediated by specific binding of the gel promoting cations (Rees et al., 1980). The first step in the gelation of carrageenan is the transition from a disordered random coil to the ordered helical state (Viebke et al., 1994).

Gellan Gum

Gelation mechanism of gellan gum is based on the domain model (Morris et al., 1996; Morris, 1998). In aqueous solution at high temperatures, gellan polymers are in a disordered single-coiled state. Cooling of the gellan sol promotes the formation of a threefold left-handed double helix, stabilized by internal hydrogen bonding (Chandrasekaran and Radha, 1995). Coil-helix conformational transition occurs in a temperature range from 30 to 50°C, depending on the ionic strength of the dispersion (Horinaka et al., 2004). After this transition, the gellan double helixes can be associated in the presence of cations to form junction zones, which can aggregate and lead to the formation of an interconnected three-dimensional gel network. During this step, the sol is converted into a gel. The gelation process is dependent on the type of cation, ionic strength, temperature, and polymer concentration (Sanderson, 1990), and the network structure of the gellan gels can also be modified.

Guar Gum

Gels are formed on cooling in the presence of salts. Molecules undergo a coil helix transition followed by aggregation of helices. Guar gum is readily soluble in cold water. It has a mannose to galactose ratio of around 2:1, and exhibits weak synergism (Sandolo et al., 2009; Robinson et al., 1982).

Xanthan Gum

Gels are obtained in the presence of electrolytes over a broad pH range and at high temperatures. Gels are formed on cooling. Xanthan and polymannan chains associate following the xanthan coil-helix transition (Katzbauer, 1998; Sworn, 2000).

Locust Bean Gum

Gels are formed on cooling. Polymannan chains associate following the coil-helix transition. For locust bean gum, the galactose deficient regions are involved in the association (Richardson et al., 1998; Schorsch et al., 1997).

MULTI-COMPONENT/MIXED GELS

The concept of multi-component or mixed gels arises from the application point of view. For example, the use of a single gelling agent may provide a gel that may have certain limitations like low cohesiveness, weak mechanical structural integrity, an insufficient stability towards processing and environmental conditions, unacceptable appearance, low shelf-life, and significant syneresis. Thus, mixed gel is a combination of biopolymers with inclusion of different solutes (Aguilera and Rademacher, 2004). More than one gelling agent is being used to form mixed gel (Table 5). Whey protein and polysaccharides are the two gelling biopolymers that are used to form mixed gels. Combinations of gelling agents are frequently used in food industry to attain either a desirable texture or to perform polysaccharide-protein (meat/dairy product) interaction while forming gel (Nussinovitch, 1997).

Precipitation or gelation can occur if the two hydrocolloids associate. Oppositely charged hydrocolloids are likely to associate and form a precipitate. Association of some stiff polysaccharide molecules like xanthan gum and carrageenan results in gel formation. In this system, polymers are held together by strong intermolecular interaction, and the texture and nature of the gel is entirely different from the single component gel. It is thus of great importance for producing new texture and new applications (Walstra, 2003). The mixed gels can be broadly classified as:

Binary/Two-Component Gel

Only two components are used for forming this type of gel. In Type I gel, the gel polymer A can modify the gelling characteristics of polymer B but without undergoing any interaction between them. For example, addition of dextran to gelatine increases the rate of formation of collagen triple helix structure and rate of development of gel network. Dextran and polyethylene glycol, due to their steric exclusion properties, are able to raise the helix coil transition temperature (Oakenfull, 1987). In type II gel, both polymers are capable of forming a gel network and they are of three types, such as,

i. Interpenetrating networks: Here two polymers interact only by mutual entanglements (Sikora et al., 2008). In reality, it is difficult to prepare due to incompatibility of unlike polymers. It might be possible to form one polymer network and then introduce the second polymer, and allow to gel within the pre-existing network, for example, use of alginates to recover and utilize waste protein.
ii. Phase separated network: Concentrated dispersions of mixed polymers (for example, proteins and polysaccharides) mostly lead to phase separation and de-mixing. Example includes mixed gels of agar and gelatin. Phase separation is indicated by sharp changes in melting point and elasticity modulus (Lorén et al., 2001; Lundin et al., 2000).
iii. Coupled network: Gelation may be possible when a favorable interaction exists between the chains of two polymers connected together by intermolecular binding. This structure arises from gel forming polymers with at least some junction zones involving chemical cross-links. Gel formation between alginate esters and gelatin has been reported. This type of synergic interaction is more typical among binary polysaccharide systems (Zasypkin et al., 1997).

Filled Gels

A filled gel is a gel matrix with particulate inclusions. The fillers may be gas bubbles, liquid droplets, crystals, fat, or cellular components such as starch granules. The properties of fillers change during processing. The effect of the filler on the elastic properties of the gel (reinforcement) can be quantitatively described as the ratio of the modulus of filled gel to that of the corresponding unfilled gel. Starch gel is the natural filled mixed gel, in which matrix formed by amylose and the swollen amylopectin granules act as fillers (Steeneken and Woortman, 2009).

Polysaccharides with Surfactant Micelles

An anionic surfactant (such as SDS) when added at a concentration above the CMC (critical micellization concentration), micelles are formed and interact with the polymer. Sometimes, a few polymer chains overlap with the micelles and form cross-linkages. If the polymer concentration is lesser than the chain overlap concentration, then mainly intermolecular junctions are formed, and vice versa, viscoelastic gels are developed (Lindman, 1993).

Table 5 Characteristics and application of multi-component gel

Component used	Other ingredient	Application	References
Gellan gum Sodium alginate, Carboxymethylcellulose	Soy protein isolate, chickpea flour, refined groundnut oil, common salt	Gel for reducing oil uptake in sev, a legume based product during deep-fat frying	Bajaj and Singhal (2007); Bayarri et al. (2009)
Deacylated gellan gum powder	Glucono-δ lactone (GDL), calcium, sodium, magnesium, potassium & phosphorous sources	Acid gelation of gellan: Effect of final pH and heat treatment conditions	Yamamoto and Cunha (2007)
κ-Carrageenan	Casein micelles	Aggregation of casein micelles and k-carrageenan in reconstituted skim milk	Ji et al. (2008)
Chitosan	Chicken salt-soluble proteins	Use of chitosan on physico-chemical properties of chicken salt-soluble protein gel	Kachanechai et al. (2008)
Gellan gum	Gelatin	Gellan gel-based microcarrier delivery system	Wang et al. (2008)
Gellan gum	Konjac glucomannan	Characterization of konjac glucomannan–gellan gum blend films and their suitability for release of nisin incorporated product	Xu et al. (2007)
Gellan gum	Calcium salt, azathioprins	Multiparticulate gel formulation for colonic delivery	Singh and Kim (2007)
κ-Carrageenan and gellan gum	Sucrose and aspartame	Sweet gels	Bayarri et al. (2007)
Gellan gum	β-lactoglobulin-coated lipid droplets	Protein-coated lipid droplets onto gellan gum hydrogel	Santipanichwong et al. (2008)
Carboxymethyl cellulose (CMC)	Milk	Milk based gels	Bayarri et al. (2007)
Carrageenan, gellan gum, locust bean gum	Pectin	Whey protein-polysaccharide cold-set gels	Jong et al. (2009)
κ-carrageenan	Locust bean gum	Synergistic locust bean gum-κ-carrageenan gels	Arda et al. (2009)
Gellan gum, whey protein isolate (WPI)	Glucono-δ-lactone	fracturing composites Cold gelation products	Berg et al. (2007)
Carrageenan, whey protein isolate	Soybean oil	Emulsion gel	Singh et al. (2003)
Gellan, agar	Sodium citrate, calcium chloride, clarithromycin	Gel for treating gastric ulcers	Rajinikanth and Mishra (2008)
Chitosan, whey protein concentrate	Canola oil	Emulsion gel	Laplante et al. (2006)
Chitosan	Heparin	Pharmaceutical microemulsion gel	Andersson and Lofroth (2003)
Milk	Whey protein	Protein gels	Fertsch et al. (2003)
Iota –carrageenan	Ethyl hexanoate	Flavour releasing polysaccharide gels	Juteau et al. (2004)
Starch	Ethyl hexanoate	Flavour release from polysaccharide gels	Juteau et al. (2004)
Pectin	Mixture of enzymes	Enzymes for improved gel strength	Budolfsen and Nielsen (1999)
Alginate	Calcium salt, food acids, antioxidants	Quick set desert mix gel	Hembling et al. (1995)
Gellan gum	Glycerol, and ascorbic, citric, acetic, oxalic acids, hydrogen peroxide, sodium acetate, magnesium perchlorate, sodium bromide, magnesium, chloride	Gum films as carriers of ascorbic acid	Leon et al. (2008)

Polymers with Heat-Setting Proteins

Any globular protein at a pH higher than its isoelectric point when heated can form a gel. If the same proportion of anionic polysaccharide (e.g., carrageenan) and protein is mixed well, a homogeneous dispersion is formed which upon heating form a gel. This gel shows higher elastic modulus than that of protein alone (Schorsch et al., 2000).

Starch-Reinforced Protein Gels

Starch is widely used as fillers in commercial products to strengthen the mechanical properties of gels as well as to reduce cost. The role of starch granules is to withdraw water from the system as they swell and imbibe water during gelatinization. The net result is that the effective concentration of the protein dispersion increases and when it gels at a higher temperature (e.g., 75°C for whey proteins), a strong protein matrix is formed around gelatinized starch (Aguilera and Baffico, 1997).

Emulsion Gels (Copolymer Gels)

Gels containing protein or polysaccharide-covered fat droplets exhibit enhanced stiffness if the adsorbed protein or polysaccharide outer layer interacts with the gel matrix (Sosa-Herrera et al., 2008; Aguilera and Kessler, 1989). Logaraaj et al. (2007) has developed an emulsion-gel by forming an emulsion with edible oil at the beginning followed by heat setting with soy protein to attain a well-set gel.

Instant Gel

Multicomponent instant gel is a relatively new idea wherein a gel with acceptable textural attributes is formed quickly (say, 5 min.). The factors that decide the formation of an instant gel include appropriate selection of raw materials, the nature and presence of catalyzing agents, and environmental conditions including the pH and the temperature.

Rheological behavior and microstructure of pea protein/k-carrageenan/starch gels with different setting conditions was investigated by Nunes et al. (2006). The results show that gel setting conditions had a significant influence on the phase separation process of pea protein and k-carrageenan, and hence on the final structure and textural properties of the mixed gel. Depypere et al. (2003) investigated the mixture design approach on the dynamic rheological and uniaxial compression behavior of milk desserts. Mixture design experiments were used to study the interactions between k-carrageenan, native maize starch, and milk proteins in the formation of a mixed gel structure. The milk dessert is considered as a three-component system in which the interactions between two ingredients are not isolated from the third constituent. Lee et al. (2003) characterized gellan-gelatin gels and observed that the hardness decreases with increasing gelatin proportion and cohesiveness increases upto the gellan to gelatin ratio of 40: 60 and then decreases. The concentration of NaCl and gellan to gelatin ratio are critical factors for the characteristics of the formed gel. The effect of different additives on textural attributes of multi-component gels is expected to generate restructured products like fruit-based gels.

CONCLUSIONS

Gels are viscoelastic materials, and the stability of food gels is of utmost importance for commercial use. Thus, the understanding of the conversion of sol to gel is essential for developing gelled products. A large number of factors such as the nature of material, concentration, time, temperature, pH, etc., can affect the process of gel formation, and consequently, its texture, the most critical factor for consumer acceptance. The role of hydrocolloids and proteins, and the presence of cations also dictate the structure of gel. The detailed viscoelastic properties of alginate gels can be determined by employing fundamental rheological tests such as low-strain compression, creep-recovery, oscillation, and stress relaxation. The phenomenon of strain hardening and the fracture behavior can be correlated with gel forming conditions.

There is a lack in knowledge concerning the relationship between the gel forming variables and the textural attributes of the developed gel. The gravity induced flow and spreadability, and sensory attributes of these gel products are yet to be ascertained, though these indices are mandatory to know for their wide use. New types of gelled products like multi-component or mixed gels, aerated gels, and emulsion gels are the new directions that are expected to have a good future soon. The application of these findings lie in developing restructured foods and new types of foods having adequate mechanical integrity, long shelf-life, nutritional status, and desirable consumer acceptability.

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