



Scientific Paper

Edible oleogels in molecular gastronomy

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Abstract

Experimental chefs and researchers have limited options when structuring lipid-based materials present in foods to include: liquids, solids, foams or emulsions. However, the application of gel technology for lipids is on the cusp of advancing into experimental culinary kitchens around the world. The possibility of utilizing edible oils (and even ethanol) to extract a hydrophobic flavor and then gel the material in a similar fashion as hydrocolloids gel water is now a reality. This review covers the three primary oleogels: ethyl cellulose, mixtures of γ -oryzanol and β -sitosterol and candelilla wax.
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Oleogels in foods

Modifying the physical state of foods has been a cornerstone of molecular gastronomy since its origins, and the ability to control and manipulate aqueous based systems has led to the creation of a gamut of materials including: solids, gels, liquids, and foams. When a component of a dish or food is primarily water, numerous strategies are well understood and in place to modify its physical state. It may be converted to a solid by simply cooling or evaporating a portion of the water, it can be modified to foam by whipping with air, CO₂

or N₂, or it can be gelled with one of the plethora of polymeric gelators. Polymeric hydrogelators allow complete control of the textural properties and in numerous cases allow the modification of the flavor release profile (Vilgis, 2012). Most typically, polymeric gelators, in aqueous solutions solidify as they are cooled and hence the state transition is an enthalpy-controlled reaction. This is the broadest group of gelators (i.e., gelation, agar-agar, starch, guar gum, locust bean gum, etc.) available to modernist chefs and is thoroughly reviewed by Martin Lersch (Lersch, 2010). The second strategy, also an enthalpy driven reaction, is to gel aqueous solutions by physically cross-linking the polymers using salts via the formation of salt-bridges (i.e., alginates and carrageenan) (Vega and Castells, 2012; Lee and Rogers, 2012; Campo et al., 2009). Finally, gels can also be driven to form via entropy driven state transitions whereby the gel forms at elevated temperatures and the reversible solution forms upon cooling (i.e., methyl cellulose) (Kato et al., 1978; de Moura et al., 2009). Unfortunately, these hydrocolloid based gelators are not soluble in hydrophobic liquids such as vegetable oils limiting their applications to aqueous systems.

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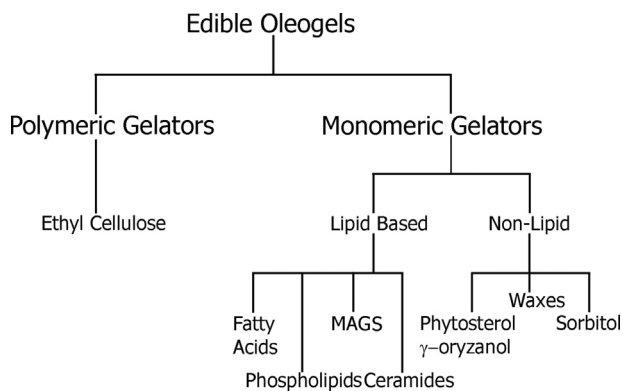


Fig. 1. Potential ingredient technologies to form oleogels.

Lipids are a major constituent of many dishes, because they provide desirable mouth-feel, flavor and impart solid-like properties to the food. Traditionally, processed foods, which contain high quantities of lipids also contain hard stock or modified hard stock fats consisting primarily of *trans* and saturated fats (Wassell and Young, 2007; Wassell et al., 2010). The macrostructure of fats and oils, including their viscoelastic properties, are highly influenced by both the nanoscale (triglyceride (TAG) composition and polymorphic form) and microstructural elements (fat crystal size and amount of solids) of the food (Awad et al., 2004; Tang and Marangoni, 2006c; Wesdorp et al., 2005). The elastic properties of lipids originate from the presence of long, straight-chain fatty acids. Upon cooling, the solubility of the hard stock TAGs decrease leading to a decrease in solubility causing phase separation and eventually nucleation followed by crystal growth. Since TAGs are large, anisometric molecules, numerous configurations in the solid states exist giving rise to three primary polymorphic forms: α , β' , and β . Depending on the desired material properties, mass and energy transfer can be modified selecting the desired form. The polymorphic arrangements stack TAGs into lamella within the crystals and it is these crystals that form a 3D network of clusters interacting via non-covalent interactions forming the final macroscopic network (Tang and Marangoni, 2006c, 2006a, 2006b). The major advantage of TAG structuring is the diversity of fatty acids and their possible molecular configurations, which allows for the physical properties to be tailored by changing the chemistry of the molecules or by modifying the processing conditions.

However, the ability to modify the structure of oils, in molecular gastronomy, is limited to changing the amount of solid hard stock fats, whipping in gas or generating emulsions. The ability to gel oil has yet to emerge as a viable technique in molecular gastronomy although numerous ingredient technologies exist (Fig. 1) (Rogers, 2009; Bot et al., 2009b; Perneti et al., 2007; Hughes et al., 2009). Herein we will report on three oleogels, ethyl cellulose, mixtures of β -sitosterol and γ -oryzanol and wax – specifically candelilla wax. Lipid based gelators, as well as sorbitol, structure oils in a very similar fashion to hard stock TAGs and as such will not be reviewed.

Polymeric gelators

Few polymeric oleogels exist due to the poor solubility of these compounds in edible oils. However, one extremely effective polymeric oleogelator has been identified – ethylcellulose (EC) (Laredo et al., 2011b; Marangoni, 2013). EC can create a solid elastic gel in oil (Co and Marangoni, 2012). EC is a linear polymer of 1,4- β -D-glucose units with ethoxy substitutions at carbons 2, 3, or 6 (Fig. 2A). It is synthesized from cellulose and when the degree of substitution reaches 2.3–2.6 (out of 3) the EC becomes soluble in organic oils. It has been found that an oleogel (Fig. 2B) can be formed when EC is heated in vegetable oil above its glass transition temperature at approximately 140 °C and subsequently cooled (Dey et al., 2011). The EC is able to partially solubilize in the oil, and once cooled, the EC polymers interact with each other and the oil, forming a gel network (Laredo et al., 2011a). These EC oleogels have been researched for their potential use in a variety of food products ranging from pure vegetable oils to heat-resistant chocolate (Stortz and Marangoni, 2013). This method has several shortcomings (Gravelle et al., 2012). A method was developed by Gravelle et al. (2012), which uses a convection oven with an overhead stirrer which offers the benefits of good temperature regulation leading to repeatable sample preparation times, minimal sample bubbling and the elimination of any granular particles. The requirement for EC to be heated above its glass transition temperature in the oil during gel manufacture leads to concerns about oil oxidation. Furthermore, any surfactants that are added to the system may be affected by the high heat and the EC is also known to start decomposing at temperatures above 170 °C (Gravelle et al., 2012). Therefore, the extent of oxidation and degradation of the system were studied during the manufacture of gels using the oven method.

It was found that the peroxide value (PV) and thiobarbituric acid, reactive substances in canola oil, increased over time while heated in the oven at temperatures above 140 °C (Gravelle et al., 2012). A holding time of 20 min was found to be the maximum heating time before the oil exceeded quality parameters for “fresh” oil (Codex Stan, 1981). Interestingly, it was found that gels made with a combination of fresh and rancid (PV > 10 meq/kg) canola oil displayed increasing mechanical strength (tested by back extrusion) with increasing proportions of rancid oil. This result was corroborated by measuring the mechanical strength of EC oleogels held for various amounts of time in the oven after reaching 140 °C. An unexpectedly large increase in mechanical strength was observed between 5–10 min holding time, which was attributed to dissolution of more of the EC rather than just an effect of increased PV. The increasing gel mechanical strength with increased PV can be attributed to the greater polarity of the oxidized oil that would increase the solubility of the EC in the oil leading to a stronger gel matrix.

Depending on the desired physical properties and the food material of interest vastly different gels may be required. Therefore, the properties of EC gels were studied using an array of formulation variables. EC is commercially available in

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