



A colloidal gel perspective for understanding oleogelation

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The field of oleogelation have shown a tremendous progress in the last decade both in terms of fundamental exploration as well as practical applications. However, one of the main bottleneck that still limits the full-scale commercial exploitation of oleogelation techniques is identification of ideal oleogelator (s) with desired properties. Most, if not all oleogelators that are currently been explored in the field, have been identified serendipitously. A rational understanding of the structuring mechanisms and the consequent gel properties of existing systems could serve as a catalyst to accelerate our efforts in finding the ideal oil gelling agents. The purpose of this opinion paper is to look at oleogelation from a colloidal gel perspective in order to have new insights into the gelling properties of different categories of oleogelators forming mono and multicomponent gels.

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Introduction

Academic research in the area of oleogelation has progressed rapidly as researchers from wide range of backgrounds (such as applied chemistry, colloid science, material science and process engineering) have taken-up the challenge to identify novel ingredients and innovative processing ways to create oleogels. Fundamentally, it is quite fascinating to fabricate and characterize these new class of edible soft matter systems as there is still a lot to be learnt about their structuring mechanisms and ‘tunability’ of their bulk properties through microstructural alterations. And from application point-of-view,

oleogelation approach has potential to cater to a number of generalized and ‘niche’ applications.

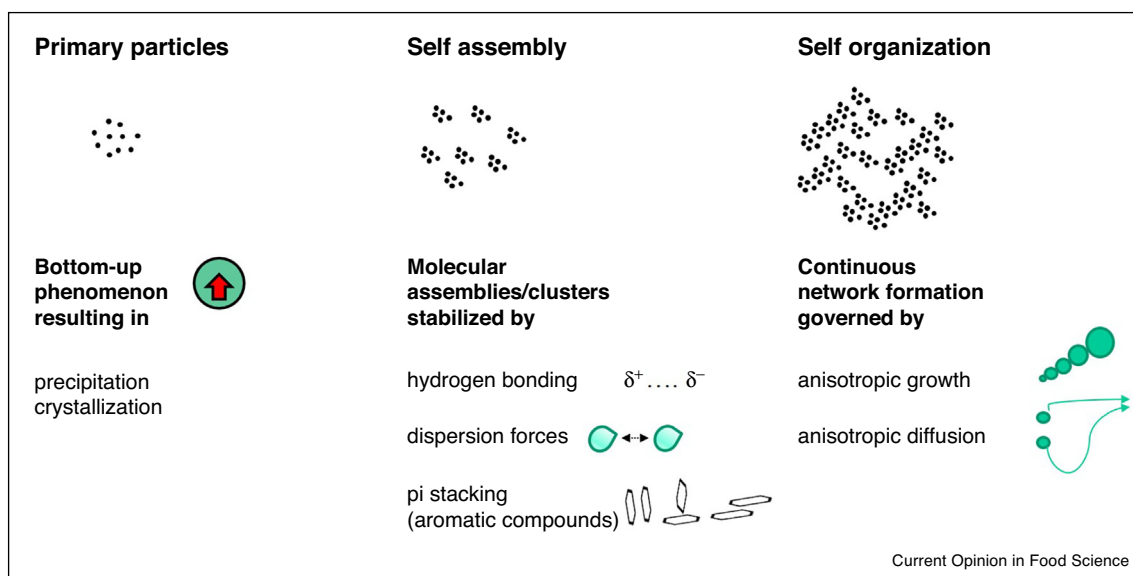
Oleogelation and monocomponent gels

One of the main bottlenecks in the field of oleogelation is finding structuring agents (oleogelators) that are effective at low concentrations, cheap, readily available and most importantly, have required regulatory approval for use in edible products [1,2*]. Mechanistically, the most important requirement for a material to act as an oleogelator is to show a suitable balance between their affinity for edible oils (*i.e.* weak interactions with unsaturated triacylglycerols) and sufficient insolubility in these solvents in order to trigger molecular self-assembly (process by which individual molecules form defined aggregates) and subsequent self-organization (process by which the aggregates create higher-ordered structures) as a function of supersaturation and external factors such as temperature that can alter the solute–solvent interactions. Higher-ordered structures such as crystal lattice, liquid crystals, micelles, bilayers, fibrils and agglomerates, may result in the formation of a 3-dimensional network that can physically trap liquid oil and lead to oil gelation. These molecular assemblies or building blocks are usually stabilized by non-covalent interactions such as hydrogen bonding, short-range van der Waals attractive interactions (dispersion forces) and π – π stacking. These non-permanent or transient bonds are characterized by a finite lifetime so they can reversibly break and form many times during the course of an experiment. The growth of molecular clusters or assemblies into continuous network is further dictated by anisotropy of specific surface free energy (at active sites) as well as the anisotropy of the mobility of diffusing molecules (or kinetic coefficient) [3]. Anisotropy of specific surface free energy (at active sites) is the difference in the local surface free energy at active sites (growth sites) compared to non-active sites in a growing crystals or clusters. This gradient in surface free energy determines the orientation of incoming molecules and the equilibrium growth shape which is aimed at minimization of surface free energy. In addition to this local factor, growth rate and equilibrium growth shape is also affected by diffusional or long-range influences. The process of gel formation is depicted schematically in [Figure 1](#).

Mechanism(s) involved in oleogelation

Most work reported on edible oleogels in the literature have only lightly touched upon the mechanism responsible for the formation of the structuring units and gel formation. Knowledge related to self-assembly and self-organization of oleogelators have been mainly obtained

Figure 1



Infographic representation of stages involved in gel formation. Weak solute (gelator)–solvent interaction triggers formation of primary particles through precipitation and/or crystallization ('bottom-up' phenomenon) followed by self-assembly and self-organization to form higher ordered supramolecular structures, that eventually leads to gelation of solvent if a proper balance between gelator–gelator and gelator–solvent interactions is achieved. These interactions are influenced by anisotropic growth of supramolecular structures and anisotropy in diffusion kinetics of molecules or clusters of molecules.

from in-depth studies done using 12-hydroxystearic acid and its derivatives as gelators in a range of organic solvents [4,5]. A correlation of self-assembled structure formation with the solvent properties (Hansen solubility parameters) have also be explored to better understand the mechanism involved in the formation of supramolecular gels [6^{••},7–9]. In general, a weak solvent–gelator interaction results in a dominant gelator–gelator interactions which may lead to the formation of continuous network. However, a much stronger gelator–gelator interaction will eventually lead to precipitation of crystalline or amorphous molecule clusters (phase separation). Hence, an optimum balance between solvent–gelator and gelator–gelator interactions ultimately governs the formation of a continuous network (percolated network) of self-assembled gelator molecules and subsequent gelation of the solvent. As seen with the colloidal gels [10[•]], arrested phase separation can be considered as one of the mechanisms that drives 'out-of-equilibrium' gelation in oleogels. Basically, the micro to macrophase separation is interrupted by dynamical arrest leading to gelation of solvent. Macrophase separation is an equilibrium state where the two phases show complete phase separation. Microphase separation on the other hand, is a non-equilibrium state where colloid-rich micro domains are formed which may be random or follow a regular pattern.

In other cases, equilibrium gelation may follow the conventional route which involves particles forming transient

clusters which in turn form transient network that transform further into percolation of long-lived clusters, eventually resulting in gels. Thus, the gelation process is rather complex as it involves supramolecular interactions at primary, secondary and even tertiary levels.

Considering these mechanisms, some of the important features that are required for gelation include: (a) strong directional intermolecular interactions of gelator molecules that promote unidirectional growth of supramolecular aggregates; (b) ability to form inter-network secondary interactions leading to intertwined aggregates, (c) factors preventing neat crystallization of gelator molecules, and (d) limited re-organisation of mesoscale structure after network formation [11^{••}]. Accordingly based on this understanding, molecular and crystal engineering concepts could be utilized to design and identify new gelator molecules. For instance, molecular features such as having moieties with hydrogen bonding functionalities and long alkyl chains which are capable of self-assembling (via van der Waals interactions) could provide early indication of gelation properties based on molecular structures. On the other hand, crystallization properties such as anisotropic interactions that promote growth of unidirectional crystals and retardation of growth in other two directions (leading to suppression of lateral growth) or minimization of post-crystallization events such as crystal aggregation, may result in a more 'spread-out' crystalline phase in the continuous phase resulting in the formation

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