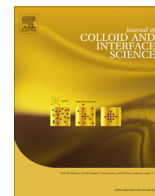




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Structuring of colloidal particles at interfaces and the relationship to food emulsion and foam stability

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ABSTRACT

We consider the influence of spherical colloidal particles on the structure and stabilization of dispersions, emulsions and foams. Emphasis is placed on developments in the use of the methods of liquid state theory and computer simulation to understand short-range structuring of concentrated colloidal dispersions and ordering of particle layers near surfaces and within liquid films. Experimental information on the structuring of surfactant micelles and caseinate particles in thin liquid films is described, including an assessment of the effect of particle polydispersity on depletion interactions and kinetic structural stabilization. We specifically discuss the relevance of some of these structural concepts to the stability of food colloids.

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1. Introduction

For budding investigators aspiring to establish a body of work capable of recognition by their academic peers as both rigorous and significant, the fields of food science and food technology might be considered unpromising areas of research. Pragmatic justification for this view would have its origin in the well-founded perception of the ‘messiness’ of food systems, which presumably conspires to inhibit the systematic planning and execution of traditional hypothesis-driven experimental investigation and associated theoretical analysis.

One way to try to get around the problem of the compositional and structural complexity of foods is to adopt a strongly reductionist

approach. This involves the specification and systematic investigation of much simpler model systems that are perceived to contain the key structural attributes or essential functional ingredients of real food products. One then argues intuitively [1] that the physico-chemical behaviour of the real food product is essentially similar in certain crucial respects to the well-chosen model system. A successful exposition of this approach is to be found in the research work of Professor Darsh Wasan and his colleagues on the structuring of colloidal particles in thin films, foams and emulsions [2–5].

The outstanding stabilizing properties of familiar natural ingredients such as casein, gelatin, albumin and gum arabic were recorded by the pioneering colloid scientists. According to one of the earliest quantitative measures of the ability to act as a ‘protective agent’, published back in 1901 [6], the food proteins casein and gelatin were assigned low values of the so-called ‘gold number’

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(Goldzahl) in recognition of their impressive effectiveness as colloid stabilizers. In contrast, other natural polymers like starch and dextrin ('non-stabilizers') were placed at the opposite end of the scale. Based on such information, the essential principles of colloid science were gradually established within the physical chemistry community in the early years of the last century. Nevertheless, the relevance of colloid science to food technology was not properly recognized until after the Second World War. Of particular importance in this connection was a pioneering paper published by Hauser [7] in the month of this author's birth (April 1948) entitled "The Importance of Colloid Chemistry in Food Technology". Hauser's wide-ranging article (including a record of lively symposium-based discussion) contains straightforward explanations of the basic colloid science principles underlying the main areas of food production. In particular, attention is directed towards the putative role of functional ingredients involved in the formulation and stabilization of a familiar culinary emulsion—mayonnaise. Hauser's article also contains a quotation from a certain Mr. J.J. Corran, the then Chief Chemist of J. & J. Colman Ltd (UK), which seems relevant today: "A complete understanding of the digestion of our food, particularly of fats, is dependent, amongst other things, on a knowledge of emulsions." Indeed, the investigation of the stability properties of emulsions, both before and after eating, does still remain a vibrant area of research activity within the food colloids community.

The year 1948 could also be regarded as the start of the modern colloid science era with the publication of the classic treatise of Verwey and Overbeek on the stability of lyophobic colloids [8]. This monograph describes how two independently derived concepts—interacting electrical double-layers and van der Waals dispersion forces—can be combined into a general theory of colloid stability (the DLVO theory) which is both rigorous and predictive [8]. Unfortunately, however, in the field of food colloids we have come to recognize that the range of usefulness of the DLVO theory is rather limited [1]. One notable situation of successful applicability, though, is the predicted ionic strength dependence of the flocculation stability of model emulsions prepared with globular proteins [9]. But the failure of the DLVO theory more broadly in the food colloid arena is a consequence of the molecular and structural complexity of food ingredients. That is, multicomponent mixtures of adsorbed biopolymers and particles are typically involved in food emulsion stabilization, and their combined presence at interfaces leads to a synergy of polyelectrolyte and steric contributions to the overall droplet-droplet interactions. In these complex multicomponent systems, the structure and dynamics of the stabilizing interfacial layer is affected by a range of molecular phenomena such as complexation, self-assembly and coacervation, as well as by the competitive adsorption of biopolymers and small-molecule surfactants [10–15].

A further complication is that the properties of food emulsions (and foams) are affected also by the presence of *non-adsorbed* macromolecules and particles located in the continuous medium between pairs of dispersed oil droplets (or gas bubbles). The attractive interdroplet forces induced by the presence of these non-adsorbed species (protein particles, hydrocolloids, surfactant micelles) are responsible for the commonly encountered phenomenon of depletion flocculation [9,16,17]. Furthermore, the structuring of these non-adsorbed species in the narrow gap between fat droplet surfaces may have a positive stabilizing influence on food emulsions [2,4,18]. The role of the interparticle interactions on the structuring of concentrated colloidal dispersions requires a knowledge of the statistical mechanical concepts developed originally for simple liquids [19]. An influential contribution to this approach was made by Professor Wasan and his colleagues in modelling the structuring of colloidal particles at liquid interfaces, and in characterizing the essentials of particle-based structure formation in model food-related systems.

This article reviews how the principles of liquid state theory and computer simulation have proved useful for describing structuring of colloidal dispersions at surfaces and in thin films. We show how such concepts have provided mechanistic insight into the stability properties of emulsions and foams containing nanoparticles, surfactant micelles, and protein aggregates. While limitations of space require that the discussion here be restricted to the role of non-adsorbed particles, one should be aware that there exist many other situations for which the application of statistical modelling and/or computer simulation is a valuable tool for describing the properties of food colloid systems. Examples include the study of structure and rheology of aggregated particle gels [20–24], the reversible heteroflocculation of mixed emulsions [25], adsorbed layer structure and interactions of biopolymer-coated surfaces [26–28], surface rheology of globular protein layers [29,30], and the competitive displacement of proteins from interfaces by surfactants [12,31].

2. Structuring of particles in dense systems and near hard surfaces

The theoretical representation of concentrated colloidal dispersions relies on exploiting the analogy with simple liquids [19]. According to the classical theory of van der Waals, the structure of a dense liquid is determined primarily by the short-range repulsive interactions between the molecules, with the intermolecular attractive forces merely providing the uniform background potential in which the molecules move. Consequently, an infinitely repulsive particle-based system, the hard-sphere model, provides a reliable reference system for describing the properties of real

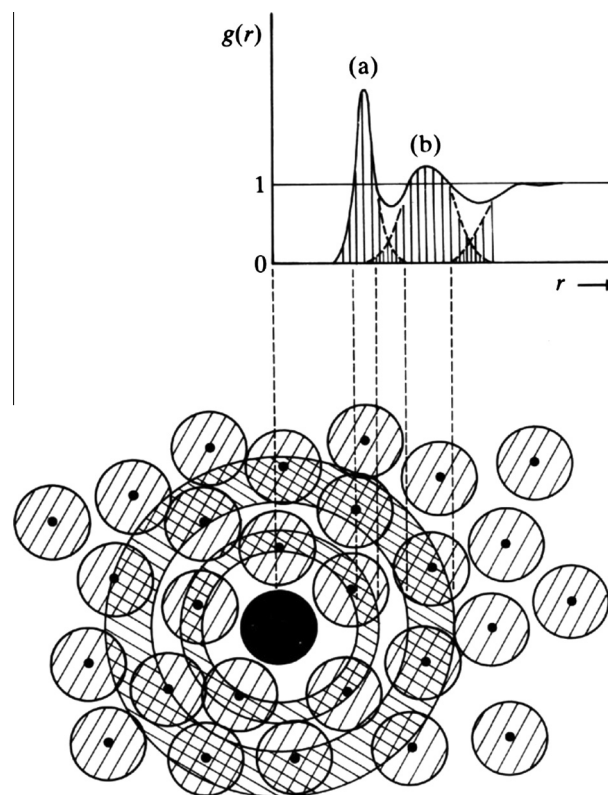


Fig. 1. Two-dimensional representation of the structure of a concentrated dispersion of spherical colloidal particles. The graph shows a plot of the radial distribution function, $g(r)$, where r is the centre-to-centre distance between a test particle and the neighbouring particles. Peaks labelled (a) and (b) indicate first and second coordination shells.

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