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Applications of ultrasonics in food science - novel control of fat crystallization and structuring

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ABSTRACT

Low power (<10 W m^{-2}) ultrasound spectroscopy has been used for many years for the characterisation of food colloids with respect to particle size distribution, adiabatic compressibility, particle solvation and dissolution, crystal nucleation and solid content. Whilst high power (>1 kW m^{-2}) ultrasound methods are well-known to impact on fat crystallization and structuring, they have many drawbacks, causing off-flavours through product oxidation and a metallic taste probably associated with sonotrode wear. Furthermore, process development with power ultrasound is hit and miss, applications being largely empirical and poorly understood. We have recently shown that well-controlled crystal nucleation can be obtained using low power, quasi-continuous ultrasound and acoustical pressure fields, opening up a new field of application in food processing for ultrasonics.

1. Introduction

The use of low power ultrasonics in food science extends over nearly forty years and in this author's case can be traced back to 1981 [1••]. Acoustical analysis of food structure including objective measures of human sensory perception is now a lively topic [2,3,4•,5•]. High power applications can be traced back to 1959 [6].

The application of ultrasound techniques to fat crystallization and structuring started with work by Miles and Fursey in Bristol [7•] which was then further developed in Leeds by the author in conjunction with Eric Dickinson and Julian McClements [8••]. Work in the area has continued continuously since then and developments are summarised in two recent papers: [9••], a recent view of fat crystallization in food colloids which is part of a journal issue dedicated to Eric Dickinson's pioneering work and [10••].

This review is mostly confined to developments in the past five years.

Ultrasound in food science may conveniently be divided into two areas [11]: low power (<10 W m^{-2}) for material characterisation and high power (>1 kW m^{-2} and >10 kW m^{-2} for cavitation in aqueous systems) for material modification and processing. However, it has recently been shown that under some circumstances, even low power ultrasound may be material altering [10••].

A recent review of ultrasound in food technology [12] gives a comprehensive, albeit uncritical, overview covering applications in filtration, freezing and crystallization, de-frosting and thawing, de-foaming, degassing, de-aeration, cutting, drying, tempering, bleaching, cooking and sterilization, extraction, mixing, de-polymerization, de-moulding, extrusion, meat tenderization, brining, pickling, marinating, emulsification, homogenization and enzyme inactivation. A better although less cited review is that by Awad et al. [11••] which also addresses low power ultrasound applications and theory. The processing effects described in these reviews are achieved mostly in an empirical manner through a range of sometimes contradictory processes associated with power ultrasound creating stable and transient cavitation [13]; free radical production and intense shear are also associated with bubble collapse in transient cavitation.

Below we consider the relationship between fat crystallization and fat structure and consider the role of both high power and low power ultrasound in the control of fat crystallization and structure.

2. The relationship between fat crystallization and fat structure

The macroscopic structure of foods containing fats such as margarine, fatty spreads, butter, mayonnaise and ice cream may be viewed as the emergence of different structures at different scales. At the smallest scale, that of individual molecule surrounded by similar molecules, all objects are in motion and without a fixed position relative to each other. Even here there is structure, for example if we take the case of water each water molecule as it tumbles around sees on average

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a structured environment with a higher density of molecules coordinating with each of the hydrogens. The crystalline unit cell, the individual crystal habit and the morphology associated with many separate crystals together form a space filling network which imparts rigidity and structure on the product. During the manufacture of a fatty spread an oil-in-water emulsion is first produced in which crystal nucleation is initiated. The transformation from the disordered liquid state to an embryonic then stable solid nucleus was recently discussed in [10••] and what appears there will not be repeated here.

Distinguishing the point at which a solid state has emerged in an oil or any saturated solution (The solid state of a pure material may be regarded as emerging from a saturated solution of itself) has created a great deal of discussion and here ultrasound has a lot to offer. The speed of sound in any material can be described by the relationship:

$$c = \sqrt{\text{Elastic constant } M \Big/ \text{Density } \rho}$$

In solid material the elastic constant comprises a bulk modulus K and a rigidity modulus G.

$$c = \sqrt{\left(K + {}^4/_3 G\right) \Big/ \rho}$$

In a fluid such as water $K \approx 10^{10}$Pa whilst even in food gels $G \approx 1 - 10$ Pa. So the speed of sound in fluids is well described by the Wood Equation

$$c = \sqrt{K/\rho} = \sqrt{1/\kappa\rho}$$

where κ is the adiabatic compressibility.

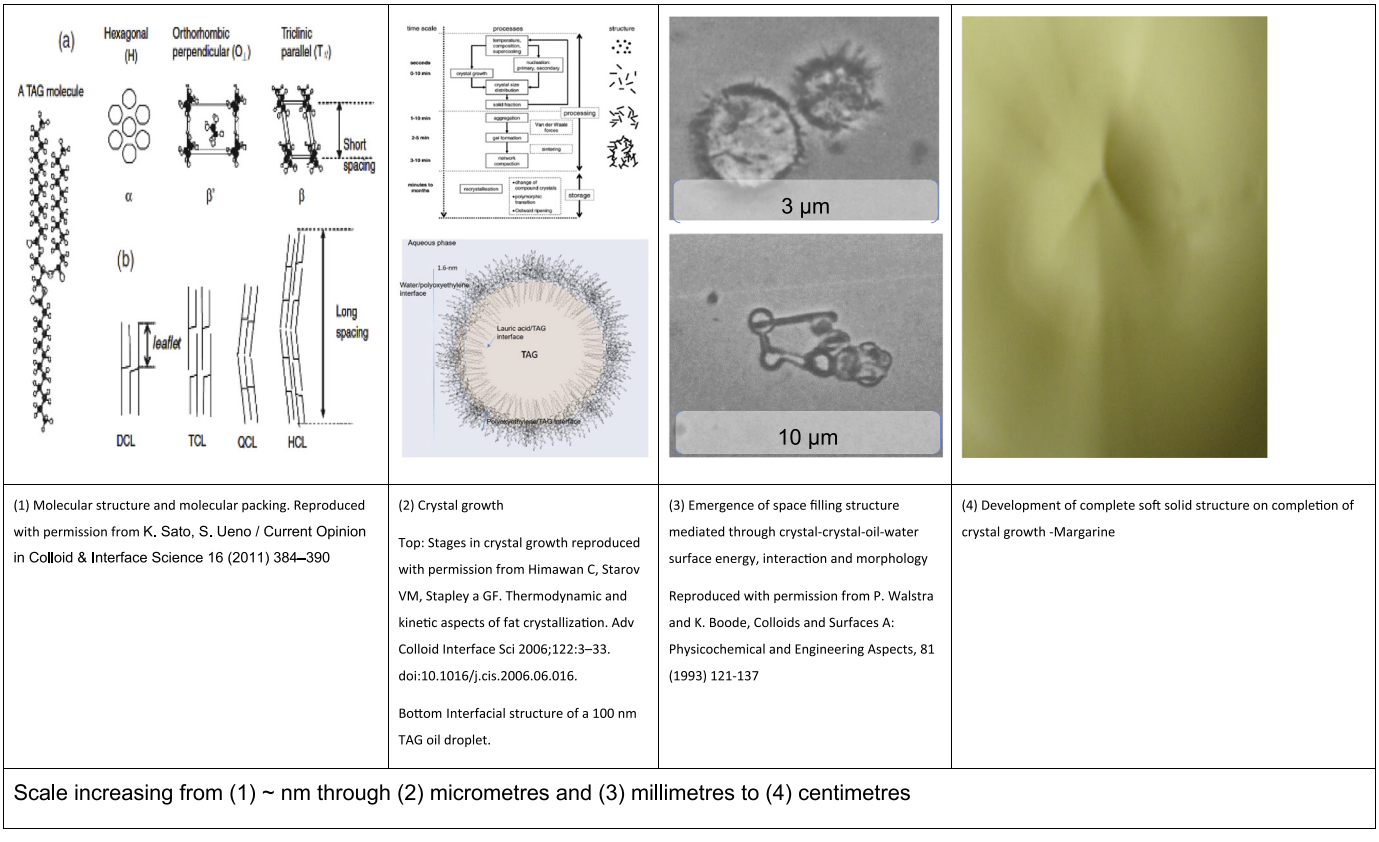
It is not only the density that undergoes a first order transition through the liquid–solid phase transition but also the adiabatic compressibility (G increases from a few Pa to the same order as K). Since the speed of sound can be measured routinely in a manufacturing process to 5 significant figures (in-line density measurements are not so easy to make and the changes in density are smaller) very accurate determinations of the initial appearance of solid nuclei from a cocoa butter melt can be made (See also Fig. 4 below). Some time ago we measured the effect of cocoa butter seed crystals on cocoa butter crystallization and the impact of these seed crystals on the chocolate tempering process is still widely ignored throughout the chocolate confectionery industry despite its impact being well-known, albeit poorly understood [14,15,16•]. Yet, removal of the seed crystals from the melt suppresses nucleation of the required Form V.

Thus far our discussion has confined itself to the first stage (1) in Fig. 1.

As shown in Stage (2) of Fig. 1 initially the crystals grow individually out of the oil droplets they begin to stick to each other (Fig. 1(3)) and the emulsion inverts as the solid structure fills space and the water becomes dispersed as small droplets within a continuous fat matrix (Fig. 1(4)), locking them away from microbial and mould growth and imparting on the resulting material a soft solid structure with desirable organoleptic properties.

In chocolate, the creation of the correct polymorph is essential to the production of a stable product with a long shelf life and attributes such as glossiness, mould release, snap, cooling in the mouth and a sharp melting point. Ultrasound offers many opportunities to study these processes at the various scales at which they occur and current methods for doing this are reviewed in Section 4.

Recently a lot of attention has been given to the stereochemistry of fats and its implications for structuring [17,18••].



(1) Molecular structure and molecular packing. Reproduced with permission from K. Sato, S. Ueno / Current Opinion in Colloid & Interface Science 16 (2011) 384–390	(2) Crystal growth Top: Stages in crystal growth reproduced with permission from Himawan C, Starov VM, Stapley a GF. Thermodynamic and kinetic aspects of fat crystallization. Adv Colloid Interface Sci 2006;122:3–33. doi:10.1016/j.cis.2006.06.016. Bottom Interfacial structure of a 100 nm TAG oil droplet.	(3) Emergence of space filling structure mediated through crystal-crystal-oil-water surface energy, interaction and morphology Reproduced with permission from P. Walstra and K. Boode, Colloids and Surfaces A: Physicochemical and Engineering Aspects, 81 (1993) 121-137	(4) Development of complete soft solid structure on completion of crystal growth -Margarine
Scale increasing from (1) ~ nm through (2) micrometres and (3) millimetres to (4) centimetres			

Fig. 1. Emergence of structure as scale increases from the molecule through a single crystal and interacting crystals to a solid structure containing liquid.