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Diffusing wave spectroscopy (DWS) methods applied to double emulsions

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Diffusing wave spectroscopy (DWS), which makes use of multiple scattered light in concentrated and highly turbid colloidal systems, has emerged as a very reliable and promising technique to probe their dynamics and rheology. However, the application of DWS to the multiple emulsion systems, particularly double emulsions (DEs), has been relatively limited so far despite their significant potential in the diverse fields of applications including food, pharmaceuticals, and cosmetics. The potential of DSW could facilitate gaining a more detailed understanding and quantitative measurement of the factors that govern the dynamics, structure, and properties of DEs, and most importantly their stability. This review briefly describes fundamental principles of DWS and its potential application to DEs to study the evolution of dynamics and structure at various time and length scales, as well as the rheological properties.

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1. Introduction

Double emulsions (DEs) can be described as ‘emulsions of emulsions’, in which both oil-in-water (O/W) and water-in-oil (W/O) emulsion exists simultaneously. There are two main types of double emulsions; water-in-oil-in-water (W/O/W) and oil-in-water-in-water (O/W/O) emulsions, the former being more commonly prepared. Interest in double emulsions stems from the desire to encapsulate and protect active ingredients in the internal emulsion droplet for pharmaceutical, food, cosmetics and biomedical application [1]. The presence of two different interfaces requires a delicate ratio of lipophilic and hydrophilic emulsifying agents for the DEs to remain stable. For stabilization of a W/O/W emulsion, a predominately lipophilic emulsifier is required to stabilize the primary W/O emulsion, the inner droplet, and a predominately hydrophilic emulsifier to stabilize the secondary O/W emulsion, the outer droplet [2**]. This is reversed for an O/W/O emulsion. Finding the ideal emulsifier combination to ensure adequate double emulsion stability is a complex task, and is most often elucidated by empirical observation of stability, without detailed understanding of how the different emulsifiers alter the oil/water interfaces in the system. Therefore quantitative measurement of the factors controlling emulsification and DEs stability hold the key to controlled emulsion properties and provide a window into the area of coalescence fine-tuning [3].

2. Double emulsions- stability and applications

DEs are deemed unstable when the emulsion droplets undergo coalescence or creaming, resulting in the ultimate phase separation of the two immiscible liquids. When two droplets are in close proximity (within the range of attractive van der Waals interactions), coalescence occurs when the intervening liquid film ruptures, allowing the droplets to merge [4]. Creaming is characterized by the separation of the fat phase of an emulsion, and ultimately phase separation, and is governed by Stokes law. Thus, the stability of a DE is dependent on a number of factors including; the selection of stabilizers or surfactants with different hydrophilic-lipophilic balance (HLB) value, droplet size, water/oil mass ratio, and viscosity of the continuous phase [5].

The destabilization of DEs is a result of one (or a combination) of four main breakdown processes (Fig. 1) which occur during storage and production [4,6–8], including:

(i) coalescence of the internal droplets;
(ii) coalescence of the outer droplets containing the internal droplets;
(iii) rupture of the film separating the internal droplets from the external phase, resulting in the expulsion of the internal droplets to the outer phase; and,
(iv) diffusive exchange between the internal droplets and the external phase through the dividing liquid film can result in the shrinkage or swelling of the internal droplets.

Emulsion systems are thermodynamically unstable due to the excess free energy associated at the surface of the emulsion droplets. Emulsifiers promote stabilization by lowering the interfacial tension and by generating strong repulsive forces between the droplets [9]. As discussed earlier, the presence of two different oil/water interfaces requires a delicate balance of emulsifying agents at the inner and outer droplet interfaces for the DE to remain stable. However, interaction between the two emulsifiers can occur as a result of migration of the inner emulsifier to the outer interface of a DE, which can compromise the stability of the DE [6,10].

Shrinkage or swelling of the internal emulsion droplets occurs as a result of the osmotic pressure gradient between the internal water droplets and the external water phase.

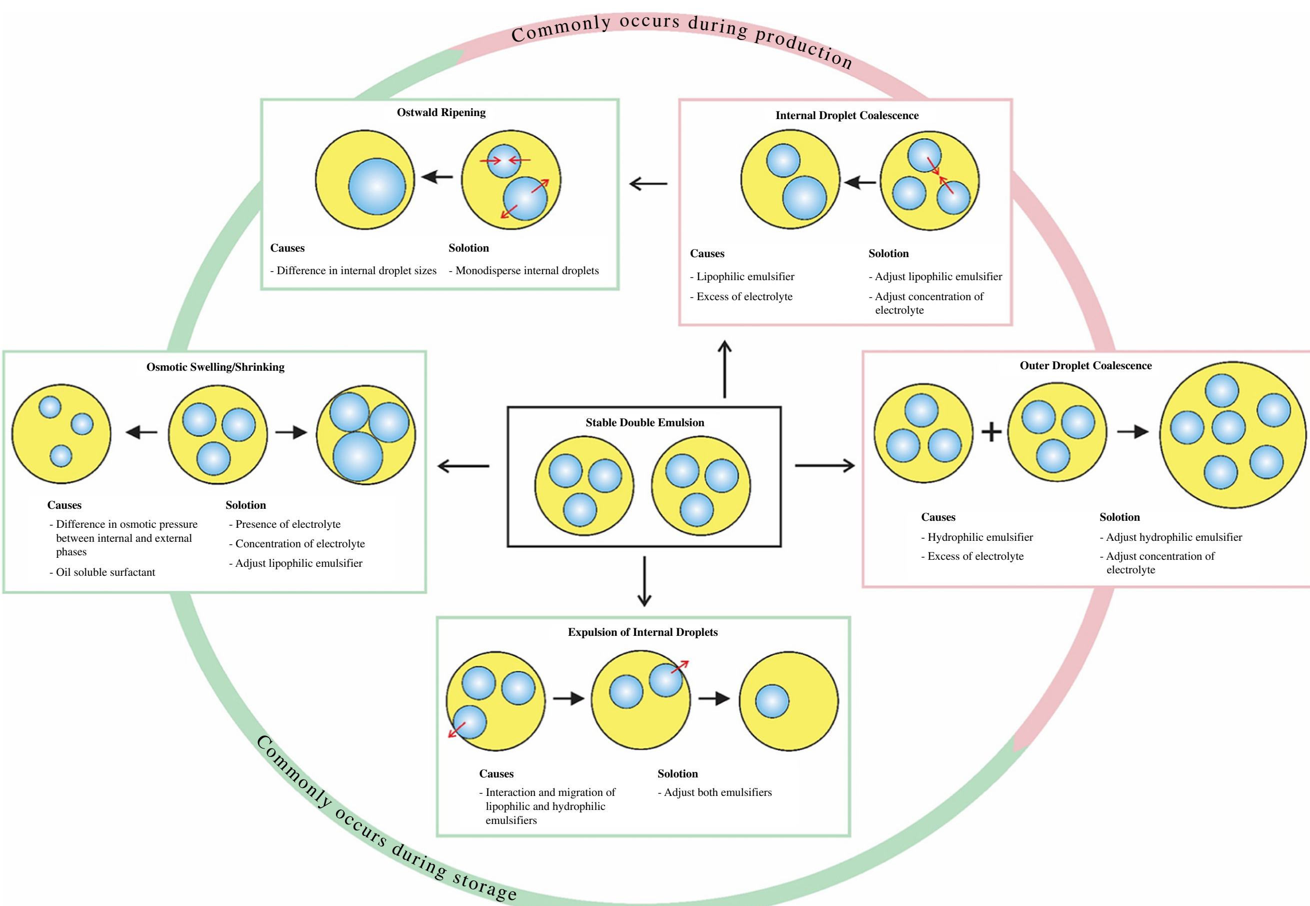

Fig. 1 Breakdown processes of double emulsions and potential solutions.