



Research Paper

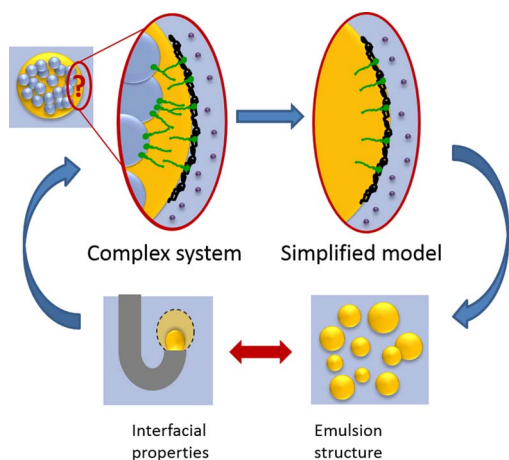
Interactions in water in oil in water double emulsions: Systematical investigations on the interfacial properties and emulsion structure of the outer oil in water emulsion



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GRAPHICAL ABSTRACT



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ABSTRACT

Due to their multicomponent structure, double emulsions are of great interest for different industrial areas. Nevertheless, there are only a few products on the market. Due to the enforced use of different stabilizing agents, production and stabilization of the double emulsions remains challenging. The stabilizing agents can interact with each other, which might cause a change of interfacial properties and emulsion structure.

To gain a deeper understanding of occurring interactions at the oil water interface, interfacial properties were measured using the pendant and oscillating drop technique. The preparation and characterization of single oil in water emulsions containing all stabilizing agents of double emulsions enables the direct correlation between interfacial properties and emulsion structure. For the interfacial and emulsification experiments, polyvinyl-alcohol (PVOH) with varying molecular weight and degree of hydrolysis, polyglycerol-polyricinoleate (PGPR) and sodium chloride (NaCl) were added step-by-step. Furthermore, stability to coalescence in emulsifying processes was evaluated using different devices: a colloid mill (CM) and high-pressure homogenization (HPH).

High molecular weight PVOHs interacted with PGPR at the interface but this did not influence the emulsion structure neither for efficient (HPH) or gentle (CM) droplet disruption processes. The low molecular weight

Abbreviations: PVOH, polyvinyl-alcohol; PGPR, polyglycerol-polyricinoleate; NaCl, sodium chloride; WOW, water in oil in water; CM, colloid mill; HPH, high pressure homogenization; NMR, nuclear magnetic resonance; DSC, differential scanning calorimetry; MW, molecular weight; DoH, degree of hydrolysis; lmw, low molecular weight; hmw, high molecular weight; DSD, droplet size distribution; INT, interfacial experiment; EMUL, emulsification experiment

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PVOH of slight hydrophobic character interacted with PGPR at the interface and this led to the breakage of the emulsions. At increased hydrophobicity of PVOH, no change in interfacial activity and no change in emulsion structure was found for the low molecular weight PVOH.

Based on these findings it is recommended to use high molecular weight emulsifiers to stabilize the outer emulsion of double emulsions. Thereby, it is not necessary to select emulsifiers with a well defined interfacial activity. However, the viscosity of the product is high in this case. If a low product viscosity is required, the hydrophilic emulsifier has to be selected carefully due to a specific interfacial activity.

1. Introduction

Due to the highly promising application possibilities for encapsulation purposes, water in oil in water (WOW) double emulsions have been in the focus of research for the last decades. Although various industrial sectors have expressed high interest in WOW emulsions, there are only few double emulsion based products on the market. The insufficient physical stability of double emulsions, caused by their complex structure, remains the biggest challenge for a successful production and marketing of multiple emulsions [1–3].

The presence of two different interfaces requires the use of at least two emulsifiers [4,5]: A hydrophilic one to stabilize the outer oil in water emulsion and a lipophilic one to stabilize the inner water in oil emulsion. Due to their amphiphilic character, these emulsifiers are expected to not exclusively adsorb at the respective interface. Particularly the lipophilic emulsifier, which is solved in the intermediate oil phase, can adsorb at both interfaces and might compete or interact with the hydrophilic emulsifier at the oil water interface. This adsorption will change the interfacial properties and may lead to a destabilization of the emulsion. Kanouni et al. [6] assumed that hydrophilic emulsifiers used in the outer water phase with high interfacial activity could displace the lipophilic emulsifier and thereby destabilize the double emulsion. The authors suggest using a less interfacial active emulsifier to avoid this destabilization. Garti and Aserin [7] showed that an efficient combination of polymeric surfactants and small molecular weight emulsifiers can improve the double emulsions' thermodynamical stability and the encapsulation efficiency.

Even though there are studies dealing with the effects at the interfaces and their implication on double emulsion structure, it is still a challenge to relate the interfacial properties directly to the emulsion structure. Most of established measurement techniques offer the opportunity to determine the interfacial properties of a single interface [8,9]. The independent behavior of two interfaces like in double emulsions cannot be characterized using these techniques. However, Won et al. [10] proposed a technique which might offer the opportunity to evaluate the interfacial properties and instability behavior of a double emulsion using a model system. Thereby, a droplet is injected into a larger one. The measurement technique is based on an adaption of the so-called oscillating drop and bubble pressure tensiometer [11].

Besides characterization of interfacial properties, the characterization of the double emulsions itself is also challenging. The established measurement techniques have high demands on the sample quality. Using nuclear magnetic resonance (NMR) diffusometry as reported by Bernewitz et al. [12] the sample has to be stable against creaming/sedimentation and thus stable against droplet coalescence. Using the differential scanning calorimetry (DSC) as described by Schuch et al. [13], the sample has to be stable during freezing and thawing procedure. In addition, using imaging or marker based techniques, interactions between added dyes or markers cannot be excluded [14]. Furthermore, the mentioned techniques do not offer the possibility to measure the superimposed instabilities like coalescence and diffusion simultaneously [15].

In this paper, we propose an approach which offers the possibility to correlate the interfacial properties directly to the emulsion structure considering double emulsion formulations. The double emulsion system was divided into the respective inner water in oil emulsion and the

outer oil in water emulsion. The present study focuses on investigations of the outer oil in water emulsion system with the aim to link interfacial properties and emulsions structure.

To the best of our knowledge, one study investigated the interactions between a hydrophilic emulsifier and the lipophilic emulsifier PGPR at the water/oil interface and the implications of the combined use on the interfacial properties and single emulsion structure. Gülseren et al. investigated the interactions between PGPR and different type of pectins at the water oil interface, and the implications on water in oil emulsion structure [16]. They reported an improved stability of the emulsion due to the building of an elastic interfacial film.

Based on our hypothesis, that interactions of lipophilic and hydrophilic emulsifiers are influenced by their molecular structure, we investigated the interfacial properties of O/W emulsions with mixed lipophilic/hydrophilic emulsifier systems varying the molecular structure of the hydrophilic emulsifier systematically. It is expected that the interactions especially the mechanism of displacement strongly depends on the interfacial activity of the hydrophilic emulsifier. Furthermore, by changing the molecular weight, the viscosity will change as well, which will influence the emulsion structure and stability. The focus of the study was to investigate whether there is an interaction between the emulsifiers and in which way this interaction plays a role for the emulsification result, especially the stabilization of the oil in water emulsion.

2. Materials and methods

2.1. Materials

Emulsions were prepared using demineralized water and rapeseed oil (Bernhard Schell GmbH Lichtenau, Germany). Sodium chloride (NaCl) with a purity > 99.5 was purchased from Carl Roth (Karlsruhe, Germany). Palsgaard (Juelsminde, Denmark) kindly supplied Polyglycerol-polyricinoleate (PGPR 4150). Different types of polyvinyl-alcohol (PVOH) were kindly provided by Kuraray Europe GmbH (Hattersheim am Main, Germany). The molecular weight (MW) and degree of hydrolysis (DoH) of the PVOHs varied for the different PVOH types. Two PVOHs with a low molecular weight (lmw-PVOH) and two PVOHs with a high molecular weight (hmv-PVOH) were selected. The PVOHs with similar MW have different DoH. The specifications of the PVOH types were:

- MW of 40 kDa and DoH of 74 mol%,
- MW of 27 kDa and DoH of 98 mol%,
- MW of 160 kDa and DoH of 88 mol%
- MW of 145 kDa and DoH of 99 mol%.

For the interfacial experiments, ultrapure water was used and the rapeseed oil was purified using Florisil® (Carl Roth Karlsruhe, Germany) as described in [17].

2.2. Methods

2.2.1. Characterization of interfacial properties

For the preparation of the aqueous phase, PVOH and NaCl were dissolved in ultrapure water at 85 °C for 2 h using a propeller stirrer.

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