Journal of Food Engineering 114 (2013) 158–163

Contents lists available at SciVerse ScienceDirect

Journal of Food Engineering

journal homepage: www.elsevier.com/locate/jfoodeng

Comparing droplet breakup for a high-pressure valve homogeniser and a Microfluidizer for the potential production of food-grade nanoemulsions

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ARTICLE INFO

Article history:
Received 23 April 2012
Received in revised form 24 July 2012
Accepted 10 August 2012
Available online 23 August 2012

Keywords:
Nanoemulsion
Homogenisation
Microfluidizer
Viscosity ratio
Emulsification conditions
Emulsifier type

ABSTRACT

A comparison of the emulsification performance of a high-pressure valve homogeniser (HPH) and a Microfluidizer has been carried out for a range of different oil to aqueous phase viscosity ratios, emulsifier types, pressure drops and number of passes through the chambers. It has been shown that for the same pressure drop across the two chambers, similar droplet sizes are produced (after 5 passes). Differences in droplet size were observed after a single pass, with the HPH producing larger droplets with a wider distribution of sizes. This difference can be attributed to the design of the homogenisation chambers with the HPH producing a wide distribution of shearing forces, so all of the starting emulsion does not experience the maximum stresses at each pass. Droplet size has been shown to be independent of viscosity ratio (0.1–80) for both homogenisers indicating that breakup is occurring in turbulent flow. No effect of emulsifier was observed in the Microfluidizer with SDS, Tween 20 and sodium caseinate. However, with the HPH, the droplet size reached a limiting value after 2 passes with SDS while with Tween 20 and sodium caseinate 5 passes were required indicating that coalescence occurs in the HPH but this is more effectively eliminated by SDS.

1. Introduction

There is a growing interest in the food industry in the use of sub micron droplets in order to prolong physical stability, enhance mouthfeel, provide faster flavour release and if the droplets are small enough (below 50 nm) to deliver oil soluble micronutrients and bioactives in an imperceptible way (McClements, 2011).

Nanoemulsions can be produced using high pressure devices such as a Microfluidizer (Henry, 2007) or low energy methods that exploit chemistry to cause phase inversion (Solans et al., 2005). However, there is no reported systematic study of the effect of high-pressure homogenisation devices on final droplet size and how this is affected by the viscosity of the oil and water phases and the type of emulsifier used. Emulsion formation is a dynamic process between droplet break-up and re-coalescence (Niknafs et al., 2011) which is controlled by the emulsifier type and rate at which it can coat the newly formed interface in the homogenisation device. The rate of formation of new interface depends on the hydrodynamic conditions within the break-up zone, the rate of energy dissipation, the viscosity of the two phases and the residence time in the break-up zone. The type of breakup is then a function of planar shear, elongational flow, turbulence and cavitation (Floury et al., 2004a,b; Walstra, 2005; Håkansson et al., 2011).

Typically nanoemulsions are produced either by using a high-pressure valve homogeniser (HPH) or a Microfluidizer. A HPH consists of a piston pump and a narrow gap, where the operating pressure is up to 150 MPa. Droplet break-up occurs within the region of the valve gap and in the jet after the gap. The advantage of a HPH is that it is scalable for industrial production. A Microfluidizer operates to a similar maximum pressure generated via a piston pump, and droplet break-up occurs from high turbulence and shear created by the collision of two impinging jets oriented 180° to each other (Cook and Lagace, 1985; Siddiqui et al., 2009). In order to determine the droplet break-up mechanism it is important to understand the geometry of the machines and the factors that affect energy dissipation including the volume over which the energy dissipates.

Several authors have used particle image velocimetry (PIV) on scaled models to obtain indications on the flow fields in a HPH (Innings and Trägårdh, 2007; Håkansson et al., 2011). It was shown that the flow into the valve gap is elongational with a higher velocity at the passage head wall than at the impact head wall. This acceleration dampens the turbulence and therefore the flow in a laboratory scale homogeniser gap is usually laminar (Innings and Trägårdh, 2007). A jet is formed at the exit of the gap where the majority of the energy dissipates: producing a stable and large eddy that causes the jet to become unstable and attach to a wall

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http://dx.doi.org/10.1016/j.jfoodeng.2012.08.009

(Innings and Trägårdh, 2007). The majority of the droplet break-up occurs at the outer regions of the jet, this is because the difference in velocity of the jet and the surrounding fluid produces the highest shearing forces.

A Microfluidizer consists of a small chamber, where an impinging plane is formed by the collision of two square inlet jet streams at 180° (each with 100–150 μm diameter). The region in this chamber is characterised by its fast dissipation of turbulent kinetic energy where the scale of segregation decreases rapidly (Gavi et al., 2007; Siddiqui et al., 2009). The majority of the droplet break-up occurs at the outer regions of the jets and the area that the jets impinge. The Microfluidizer's square inlet pipes create a larger surface area of shear with the surrounding fluid in the chamber. Subsequent to impingement the emulsion is forced through one exit pipe typically of 75 μm diameter, this dampens turbulence and produces elongational flow (Cook and Lagace, 1985) (Microfluidics, Belgium).

In this paper a comparison of the emulsification performance of a high-pressure valve homogeniser (HPH) and a Microfluidizer has been carried out for a range of different oil to aqueous phase viscosity ratios, emulsifier type, pressure drops and number of passes through the device.

2. Materials and methods

2.1. Materials

Silicone oil with viscosities of 0.01, 0.05 and 0.1 Pa s (product numbers 378321, 378356 & 378364 respectively, CAS 63148–62-9), Tween 20 (P7949, CAS 9005–64-5) sodium caseinate (C8654, CAS 9005–46-3), and glycerol (G7757 CAS 56–81-5) were purchased from Sigma Aldrich (UK). SDS (S/5200/53, CAS 151–21-3) was purchased from Fischer Scientific (Loughborough, UK). Double distilled water was used for the preparation of all solutions.

2.2. Emulsion preparation

Oil-in-water emulsions were produced by homogenising 10 wt.% silicone oil with 90 wt.% aqueous phase (3 wt.% Tween 20 and 0–50 wt.% glycerol). The low weight per cent of oil was selected to minimise affects of droplet collision and the mass of the emulsifier was tested to be in excess for the smallest emulsion produced in this paper.

A coarse emulsion was prepared by using a Silverson mixer at 5000 rpm for 60 s at room temperature. Prior to this work the effect of the coarse emulsion droplet size (5–30 μm) was tested and was shown to have no effect on the droplet size after high pressure emulsification. Nanoemulsions were produced by passing the coarse emulsion through an air-driven Microfluidizer fitted with a cooling tube maintained at 10 °C (M110S, Microfluidics, Newton, MA, USA) or a high pressure valve homogeniser (NS1001L PANDA, GEA Niro Soavi, Italy) for up to 15 passes from 50–150 MPa.

2.3. Particle size measurements

The particle size distribution and surface weighted mean droplet diameter, $d_{3,2}$, was measured by a Malvern Mastersizer MS2000 (Malvern, UK) with a Hydro SM manual small volume sample dispersion unit attached. The sample was diluted with double distilled water (RI = 1.33). For the smallest emulsions the size was verified against a dynamic laser system, high performance particle sizer, HPPS 5001 (Malvern, UK). Droplet size measurements were taken immediately after production of the emulsions.

The refractive index of the oils was measured using a Rudolph research refractometer J357 (New Jersey, USA).

2.4. Viscosity measurements

Viscosity measurements of selected samples were performed using a dynamic shear rheometer with vane geometry using a shear rate profile from 0.1 to 100 s^{-1}. All measurements were performed at 25 °C.

3. Results and discussion

3.1. Comparison of high pressure devices for effect of pass number and pressures

A series of oil-in-water emulsions were produced with 10 wt.% silicone oil (viscosity 0.05 Pa s), and 3 wt.% Tween 20 of the continuous phase. The low volume fraction of oil should reduce the effects of coalescence caused by droplet collision before the newly formed interface is coated with emulsifier. 3 wt.% Tween 20 was used as it is a low molecular weight emulsifier which should adsorb into the interface quickly and the large excess should minimise any effects of emulsifier depletion. The emulsions were passed through the Microfluidizer and high-pressure valve homogeniser (HPH) at 50, 100 and 150 MPa for 1–5 passes. Droplet size data is shown in Fig. 1 for the HPH and Fig. 2 for the Microfluidizer.

As can be seen from Figs. 1 and 2, increasing the pressure of homogenisation in both cases resulted in smaller droplet sizes. This is in agreement with previous studies (Leong et al., 2009; Qian and McClements, 2011; Donsì et al., 2011). From Fig. 1 it can be seen that in the Microfluidizer the minimum droplet size is achieved after one pass with pressure drops of 100 and 150 MPa, whereas, in the HPH this is not observed for any of the pressure drops until after 5 passes, and for the lowest pressure drop (50 MPa) there is evidence that further passes would have reduced the droplet size further. This difference is a consequence of the different geometries of the devices. The Microfluidizer creates a tight distribution of shearing forces around the maximum force (Cook and Lagace, 1985), whereas the HPH creates a wide distribution of forces. Thus, in the Microfluidizer with pressure drops of 100 and 150 MPa all the coarse emulsion entering the device experiences the highest shear forces, and as the experimental design used here has limited/eliminated coalescence, a tight distribution of

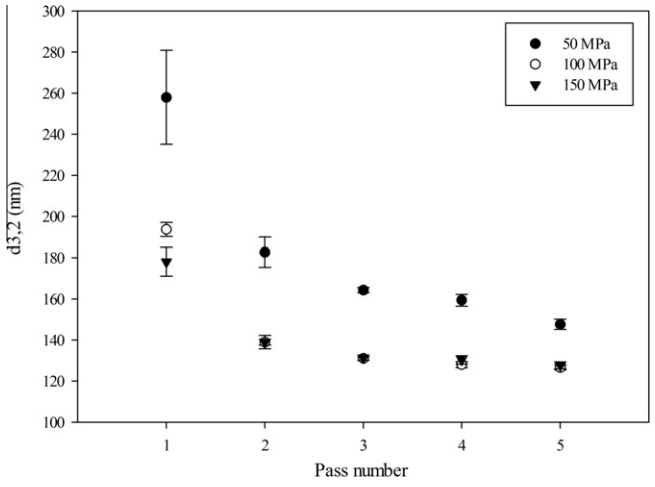

Fig. 1. Effect of pass number and pressure on 10 wt.% silicone oil (0.05 Pa s) in water emulsion droplet size with 3 wt.% Tween 20 in a valve homogeniser. Error bars for all graphs show the standard deviation calculated from three repeats.