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### Review Microfluidic emulsification in food processing

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#### A R T I C L E I N F O

#### ABSTRACT

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*Keywords:* Microfluidic systems Emulsions Monodispersity Microfluidic systems, characterized by micro-meter sized channels, are used in a variety of applications. In foods, the most common application is in the preparation of emulsions where they provide accurate control over droplet size, and shape of internal structures. This paper gives an overview of different microfluidic emulsification techniques i.e., shear driven and spontaneous droplet generation, together with their current limitations. Next a comparison is made on the basis of various parameters affecting the process of emulsification. At the end, an outlook is given on scale-up of microfluidic emulsification for preparation of food-related products.

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#### 1. Introduction

Most food products (baked goods, emulsions, frozen products, etc.) are dispersed systems that contain various components; emulsions being the most common among them (Van Dijke, 2009). An emulsion consists of droplets of one liquid suspended into another immiscible liquid. The formation and dispersion of droplets is achieved by intensive mixing or mechanical disintegration, and conventionally, the typical scale of the droplet formation

\* Corresponding author. E-mail address: abid.maan@uaf.edu.pk (A.A. Maan). unit (such as the nozzle in a high pressure homogeniser) is much larger than the droplets that are created (Skurtys and Aguilera, 2008), which implies that turbulent break-up occurs. This results not only in inefficient use of energy but also provides poor control over droplet size, shape and size distributions (Maan et al., 2011; van Dijke et al., 2010c). These features of emulsions are important to control in order to design innovative microstructures which determine various desirable characteristics of food products. Precise fabrication of emulsion droplets is possible through microfluidics which can be defined as the science of designing and operating devices consisting of channels with at least one dimension being smaller than one millimetre. These devices are aimed to deal with small amounts of fluids ranging generally from 0.001 to 1  $\mu$ L (Shah et al., 2008; Skurtys and Aguilera, 2008). The latter paper also reviews what the options of microfluidics were expected to be at the time, while the current paper reviews the latest insights.

In the last few years, microfluidic devices have gained growing attention in a variety of applications including chemistry, biology, medicine, physical sciences and energy generation (Maan et al., 2013b; Mulligan and Rothstein, 2012). In foods, the most common application is in the preparation of controlled simple and multiple emulsions with highly structured inner and outer droplets. Application of such emulsions can for example be in food drinks (improved stability due to small droplets in single emulsions), or products that may have a lower caloric load or a triggered release of flavor or other active components (double emulsions). Besides emulsification can be used to produce solid particles after phase transition of the liquid droplets, and that can be used as structural elements in food.

Microfluidic emulsification is under extensive investigation on lab scale; however, commercial application is yet not possible due to issues related to the scale up for larger throughput. In this paper, various microfluidic emulsification systems are discussed and compared in terms of parameters affecting the emulsification process, and their limitations for productivity of the overall process. Subsequently, an outlook is given on scale up of microfluidic emulsification for food preparation.

#### 2. Microfluidic emulsification systems

There is a range of microfluidic emulsification systems available. They can roughly be distinguished based on the droplet break up phenomenon. Some systems rely on shear/flow for droplet break-up and others rely on spontaneous droplet break-up.

#### 2.1. Shear based emulsification systems

In microfluidic emulsification systems, dispersed and continuous phases flow through channels, mostly as liquids that form droplets at a cross-road of channels such as in T-junctions or microchannel systems (described in next section). Alternatively, also droplets may be introduced in microchannels, to subsequently be broken up into smaller droplets while passing through a constriction, as is the case in pre-mix emulsification (Nazir et al., 2010; van der Zwan et al., 2006), or through a myriad of multiple junctions as proposed by Link et al. (2004), in which droplets are split into smaller ones. The latter two methods are not known for their monodispersity, in premix emulsification various droplet formation mechanisms occur simultaneously, while in the dropletsplitting device the manufacturing accuracy greatly influences the position at which the droplet splits, which is mostly not in the middle. In the current paper we will not consider these systems further, but focus on those techniques that are able to form very monodispersed products.

When starting from two liquids that are present in individual channels, droplets can be generated at the junction of these channels. The dispersed phase flows into the continuous phase and forms a droplet at the pore opening. In shear driven emulsification systems, the formed droplet is detached and dispersed into the continuous phase through shear exerted by the flow of continuous phase. The continuous phase flow can be in cross flowing (T-, Y-junctions; Fig. 1a and b) or co-flowing mode (co-flow systems) as can be seen in Fig. 1c and d.

Extremely monodispersed (o/w and w/o) emulsions can be produced by shear driven systems, where their surface wettability plays an important role in the type of emulsion that can be prepared (Steegmans, 2009; Steegmans et al., 2009a; van der Graaf et al., 2005). Apart from single emulsions, double and higher order multiple emulsions can also be produced by using flow focussing and co-flow systems with a controlled number and size of inner droplets (see Fig. 1e) (Hughes et al., 2013; Liang-Yin et al., 2007; Shah et al., 2008).

#### 2.2. Spontaneous emulsification systems

Spontaneous emulsification systems are characterized by two distinct channel depths; generally a shallow structure (typically called terrace) is fabricated between deeper dispersed and continuous phase channels (Kobayashi et al., 2002; Sugiura et al., 2002b; van Dijke et al., 2010d). The dispersed phase, before flowing into the continuous phase channel, is compressed on the terrace resulting in a disk like shape. As soon as disk shaped dispersed phase flows into the continuous phase channel, it spontaneously transforms into spherical droplets due to Laplace pressure differences in the dispersed phase on the terrace and in the (continuous phase) channel (Sugiura et al., 2001; van Dijke et al., 2008); see Fig. 2a–c.

Most commonly reported spontaneous emulsification systems include terrace based microchannels (Fig. 2a), straight-through microchannels (Fig. 2b) and EDGE devices (Fig. 2c). Review is available on microchannel emulsification by Maan et al. (2011) which describes various process and design parameters affecting droplet formation together with various products produced with this technique. In addition, both grooved and straight-through microchannels have been compared together with EDGE technique in terms of their emulsification efficiency. The EDGE technique is unique among microfluidic emulsification systems for its ability to produce multiple monodisperse droplets simultaneously from a single elongated pore called the plateau. The plateau is a large flat area fabricated between disperses and continuous phase channels and covered at the top with a glass plate (see Fig. 2c). Dispersed phase is pushed through dispersed phase channel onto the plateau where it spreads uniformly and forms droplets at several locations along entire length of the plateau edge. Size of the droplet is determined by plateau height with a scaling factor of around 6. Besides EDGE has been one of the very few microstructured emulsification devises used to prepare completely food grade emulsions using vegetable oil, dairy proteins, and even skim milk. Details on EDGE chip design and system set up can be found in van Dijke et al. (2010a.c).

In other microfluidic emulsification systems, including both the shear driven and spontaneous emulsification systems, droplets are produced one by one from each droplet formation unit. Only single (Kawakatsu et al., 2000, 2001b; Kobayashi et al., 2005b, 2005c, 2003; Sugiura et al., 2002c; van Dijke et al., 2010a, 2009) and double emulsions (Kawakatsu et al., 2001a; Kobayashi et al., 2005a; Sugiura et al., 2004b; van Dijke et al., 2010c) (Fig. 2d) have been reported to be produced by spontaneous emulsification systems. Higher order multiple emulsions are far from trivial due to the narrow dimensions involved in their design. Also in double emulsions, the inner droplets are reported to be produced by some traditional methods to make droplet size small enough to pass through the shallow part of the system, although in principle it should be possible to make multiple emulsions by spontaneous emulsification on one chip. Until now, the number and size of the inner emulsion droplets was found to be difficult to control in double emulsions produced by spontaneous emulsification systems, while this is easier in shear based systems.

#### 3. Comparison of microfluidic emulsification systems

Droplet size during microfluidic emulsification is primarily determined by the dimensions of the pore/nozzle. Other parameters

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