



Microfluidic emulsification devices: from micrometer insights to large-scale food emulsion production

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Microfluidic devices can be used to energy efficiently produce emulsions, and various modes of operation have been suggested in literature, and are summarized in this paper. In general, simultaneous operation of many droplet formation units in parallel is a challenge although considerable progress has been made, leading to forecast equipment dimensions that are realistic for lab-scale, and possibly full scale production. Points of attention are the inter-connectivity between the droplet formation units, and surface modification to prevent wettability changes that can be induced by the emulsifiers and stabilizers present in the emulsion during operation. Besides, for food production, microfluidics are interesting tools to investigate emulsion stability, which is expected to lead to more precise formulation and production of current food stuffs, and to *ab initio* testing of novel formulations.

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Introduction

If you mention process engineering to food technologists, the first image that will pop into their mind is a production line made of shiny stainless steel elements that are connected to make a food product or a food ingredient. Typically the dimensions of these elements are either in the centimeter range (piping, etc.) or in the meter range (unit operations). However, the essential processes that take place in order to make food or to isolate ingredients occur at a much smaller scale, typically in the micrometer, and even nanometer range. Traditional processes do not take these aspects as a starting point because these phenomena are rather inaccessible due to the turbidity of the food matrices, the small size of the structures and

mostly also the high velocities at which these processes take place.

In the last decade, extensive developments have taken place in micro- and nanotechnology (see e.g., [1] specifically for progress made in emulsion technology), and this would allow for control of processes at the scale that is most relevant to food structure formation (micrometer scale). These new technologies are in such a stage of development that various food products may benefit from the knowledge that has been gained in these fields.

In this paper, we will illustrate the latter point using emulsion production as an extensive example of how microfluidic devices can be used to investigate emulsion formation at very small scale, and these findings can be translated toward up-scaled processes, including those that can be used for derived products such as foams, capsules, double emulsions, etc. Further, we will show how microfluidic devices can help in the production of stable emulsions and the elucidation of the involved mechanisms, e.g., to measure dynamic interfacial tension at time scales that are relevant during emulsion production, and also to study emulsion stability. We will round off with how we think that microtechnology can contribute to large-scale processing, both from a viewpoint of production of foods, and as an analysis tool.

Microstructured emulsification devices

During classic homogenization processes, typically 5% of the energy that is supplied is used to make an emulsion; the rest is dissipated as heat. Since Nakashima and Shimizu filed their first Japanese patent in 1986 on the preparation of Shirazu Porous Glass (SPG) membranes, this has become the most used device for membrane emulsification; the resulting emulsions are known to be rather monodisperse [2]. In early work done at the university of Karlsruhe [3], it was quantified that membrane emulsification could be used to make emulsions much more energy efficiently, although the required membrane surfaces were still rather large in order to warrant large-scale production. This latter effect was caused by the pore size distribution in the membranes that did not allow full activation of all pores [4]. Yet even microsieves that are made through photo-lithographic techniques and that have uniform pores [5] do show pore activation that is dependent on the applied pressure [6]. It is clear that more detailed information on the droplet formation

mechanism was needed to improve the production processes, and for this microfluidic devices are the technology of choice.

Various researchers have investigated microfluidic and microstructured devices, such as T- [7,8] or Y-shaped junctions [9], flow focusing devices [10], and straight through microchannels [11,12], and good reviews have become available recently, to which we would like to refer the interested reader through to: (general reviews) [1,13^{••}]; (spontaneous droplet formation devices) [14^{••}]; (membrane emulsification for food) [15]; (pre-mix membrane emulsification) [16].

Knowledge about formation of single droplets from one droplet formation device is readily available and also various scaling relations have been suggested in literature [17[•]]. These relations work very well on the systems for which they were derived, and they accurately describe the effect of e.g., geometry and flow rates on the droplet size. A lot of good work was done in the group of Weitz and Stone (e.g., [7,10]). However, taking these findings further toward the typical scales needed for food is a major challenge.

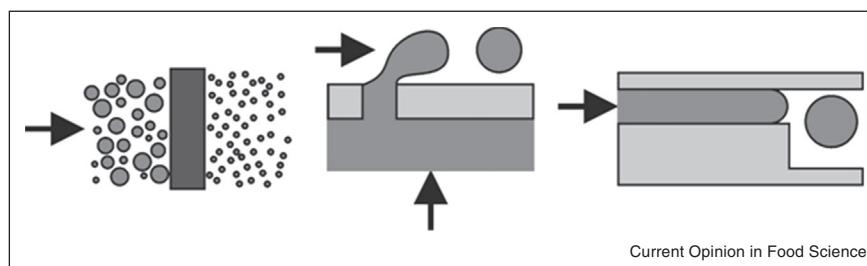
Just to name a few, the droplets that are mostly studied are orders of magnitude larger than required in food. Larger droplets are much easier to image, and that explains the preference for large droplets, but that also makes that the time scales at which the droplet formation process is studied is very different from what is needed for high throughput food production in which dynamic interfacial tension effects will occur, and which influences the droplet size dramatically as will be discussed later. Besides, the devices that are used are mostly made out of PDMS, and the liquids that are studied are model systems, and that is rather different from what would be expected from food and its ingredients. PDMS would swell in the presence of food grade oil and food components like proteins most likely exhibit less ideal

(rheological) behavior, including interactions with the surfaces. Where PDMS chips can be discarded after the experiment; this would obviously not be an option for large scale operation, and great care needs to be taken in regard to construction materials. On top of that, also the effect of multiple droplet formation units operating simultaneously in one chip is not that well known [18] and that research field would need to be developed in order to reach large scale production.

After reading all that, you may wonder why you would even consider using microfluidics, but we hope to convince you that there is still a lot to be learned. In large-scale emulsion preparation various droplet formation mechanisms may play a role, and we take these as a starting point. In general, shear-based and spontaneous droplet formation can be distinguished in microfluidic devices, and this can be carried out either in pre-mix mode, or in direct emulsification mode. Before going into up-scaling and related issues, we first explain the essence of these methods, which are schematically shown in Figure 1.

In pre-mix emulsification (see Figure 1, left), a coarse emulsion is pressurized as a whole and pushed through a porous wall, mostly a membrane. The typical droplet size that is generated is similar to the pore size, and the droplets are not extremely monodispersed. A drawback of this method is that the membrane can get blocked, e.g., by emulsifiers that are used in food such as proteins, which cannot or can only with great difficulty be removed [14,19]. In shear-based emulsification (Figure 1, middle, as an example a T-shaped junction is shown), the continuous phase exerts shear on the to-be-dispersed phase that is fed into the continuous phase through a pore. The droplet is kept to the pore by the interfacial tension but once the droplet has reached a certain size, the shear force exceeds the interfacial tension force and uniform droplets that are typically 2–10 times the pore size are formed [17[•],20,21]. To be complete, in co-flow emulsification,

Figure 1



Schematic representation of pre-mix emulsification in which a coarse emulsion is refined by passing it through a porous material (left image), direct emulsification in which the cross-flowing continuous phase shears of droplets that are formed from the oil supply channel at the bottom (middle image) and spontaneous emulsification in which the to-be-dispersed phase is constricted on a shallow part of the micro fluidic device (terrace/plateau) and that forms droplets as soon as this phase can leap into a deeper channel (right image).

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