Chapter 7

Summary

Introduction

This study was directed to develop microchannel (MC) emulsification devices for the production of monodisperse emulsions and to investigate the emulsification characteristics of the developed devices using various types of MCs and experimental systems. Monodisperse emulsions have attracted interest in both scientific and industrial fields, since they have advantages such as their better stability and simplified physical properties. Newly developed small silicon MCs with uniform sizes was employed to realize the preparation of monodisperse emulsions with micron-scale droplets by MC emulsification. A novel MC emulsification device, named a silicon straight-through MC plate, was developed to achieve higher throughput of monodisperse emulsions. This study was summarized as follows.

Summary of Each Chapter

Chapter 1

Fundamentals of microfabrication technology, microfabrication processes, and new applications of microfabricated devices in chemistry, biotechnology, and reaction engineering were reviewed. Principles, properties, and characterization of emulsions and types, fundamentals, and applications of emulsification devices were reviewed. The objectives of this thesis were also described.

Chapter 2

The preparation characteristics of oil-in-water (O/W) emulsions with micron-scale droplets by microchannel (MC) emulsification were studied using three series of silicon MCs with 1 to 3 μm-equivalent diameters. The characteristics of the prepared O/W emulsions were also studied. An open-end MC and MCs without terraces at the channel outlet exhibited unstable droplet formation with broad size distributions. MCs with terraces at the channel outlet stably yielded monodisperse O/W emulsions with micron-scale droplets; they had droplet diameters of about 5 μm and coefficients of variation below 9%. Monodisperse food-grade O/W emulsions with droplet diameters of about 4 μm were obtained by microchannel emulsification using a food-grade surfactant, polyglycerol monolaurate (PGM). The droplet size and size distribution of the recovered O/W emulsions remained almost constant over 60 d, demonstrating their long-term stability.

Chapter 3

MC emulsification and subsequent solvent evaporation were performed to prepare micron-scale lipid MS with relatively narrow size distributions. The effects of solvents, surfactants, and the lipid concentration in the dispersed phase, on MC emulsification and on solvent evaporation were investigated. The experimental systems with hexane as the dispersed-phase solvent exhibited the stable formation of oil-in-water (O/W)-MS with average diameters of 10 to 11 μm and coefficients of variation below 8% from the uniformly sized channels. Lipid MS with average diameters of 2 to 3 μm and coefficients of variation of about 20% were obtained through successful solvent evaporation of the monodisperse O/W-MS composed of 1.0 vol.% tripalmitin in hexane as the dispersed phase. The diameters of the resultant lipid MS corresponded well to their estimated diameters, regardless of the tripalmitin concentration.

Chapter 4

The formation characteristics of O/W emulsion droplets in cross-flow membrane emulsification were analyzed with real-time microscopic observations of the membrane emulsification process. A hydrophilic polycarbonate membrane with a mean pore size of 10 μm was employed. Microscopic observations of the oil droplet formation process from the membrane verified the continuous phase flow-driven droplet formation. The influences of the continuous-phase flow velocity and the surfactant type on membrane emulsification were also investigated. The use of anionic and nonionic surfactants resulted in successful membrane emulsification with no droplet coalescence at flow velocities greater than 0.1 m/s. The droplet diameter of the resulting O/W emulsions decreased with an increase in the flow velocity, remaining almost constant at flow velocities greater than 0.4 m/s. The emulsions prepared under these conditions had the average droplet diameters of about 20 μm and the coefficients of variation of 20 to 50%. In contrast, a cationic surfactant-containing system resulted in no droplet formation due to complete wetting of the membrane surface with the dispersed phase. An analysis of the surfactant-polycarbonate membrane interaction and contact angle measurements explained well the results that the membrane emulsification behavior critically depended on the type of surfactant used.

Chapter 5

We have developed a novel microfabricated emulsification device for monodisperse emulsions with high throughput of emulsion droplets. It has uniformly sized through-holes, called straight-through MC, on a silicon microchip. An oblong straight-through MC spontaneously prepared monodisperse O/W emulsions with an average droplet diameter of 32.5 μm and a coefficient of variation below 2% by forcing the dispersed phase into the continuous phase through the straight-through MC without the continuous phase flow. This successful straight-through MC emulsification was also carried out at a maximum dispersed phase flux of 65 $l/(m^2\ h)$. On the other hand, a circular straight-through MC resulted in preparing polydisperse emulsions. The results in this chapter revealed that an elongated cross-sectional shape in the oblong straight-through MC contributes significantly to the spontaneous formation of monodisperse emulsion droplets without any turbulent mixing.

Chapter 6

The effects of the device parameters and surfactant type on the straight-through MC emulsification behavior, on the resultant droplet size and size distribution were investigated. The oblong straight-through MCs with equivalent diameters of about 20 μm and elongations exceeding a threshold exhibited the spontaneous formation of monodisperse oil droplets. The unstable continuous phase flow-driven droplet formation was observed for the oblong straight-through MCs with equivalent diameters of about 55 and 160 μm regardless the value of their elongation. In contrast, the large oblong straight-through MCs with distorted cross-sectional lines and elongations exceeding three allowed the stable formation of monodisperse oil droplets with diameters of approximately 100 μm. The anionic and nonionic surfactant-containing systems used were capable of stably forming monodisperse oil droplets using an improved oblong straight-through MC. However, the cationic surfactant-containing systems resulted in unsuccessful straight-through MC emulsification. The interaction between the hydrophilic group of the surfactants and the negatively charged channel surface can explain why the surfactant charge affects the straight-through MC emulsification behavior.

The preparation characteristics of food-grade soybean oil-in-water emulsions in straight-through MC emulsification were also investigated using food-grade surfactants. Monodisperse O/W emulsions with droplet diameters of 38 to 39 μm and coefficients of variation below 3% were stably prepared from an oblong straight-through MC with a 16 μm-equivalent diameter for polyglycerol monolaurate- and polyoxyethylene (20) sorbitan monolaurate-containing systems. The rheological properties of the surfactant adsorbed oil/water interface significantly affected the straight-through MC emulsification behavior.