

Production of monodisperse *k*-Carrageenan microbeads via microchannel emulsification: Influence of flow dynamics and injection angle

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Introduction

In the past few years, droplet formation technology has found advancement with the microchannel emulsification concept (Sugiura *et al.*, 2002). In this technique, liquid extruded from a nozzle is sheared with another immiscible fluid in a microchannel, creating a monodisperse sub-micron size droplet. When compared with the conventional emulsification technique, microchannel emulsification requires lower energy input and exerts smaller amount of shearing on the droplet breakup process (Sugiura *et al.*, 2002). Therefore this technique holds great potential for bioencapsulation applications especially if the active materials are shear-sensitive.

In the past, droplet formation with a co-flowing immiscible fluid has been studied extensively in relation to the effect of process parameters, microchannel geometry and materials (Jones *et al.*, 2002; Bernhard *et al.*, 2003; Carsten *et al.*, 2004; Bernhard *et al.*, 2005; Shunji *et al.*, 2006; Darren *et al.*, 2006). It was found that emulsion with a coefficient of variation of approximately 5% and droplet sizes of 3 μm to 2 mm were successfully prepared. The production rate of emulsion was 1 to 10 ml/hr. In this work, our objective was to study the effect of nozzle insertion angle on the droplet break-up behavior, droplet size and size distribution under different flow velocities. *k*-carrageenan was used as the model dispersed phase solution and palm-oil was used as the model immiscible continuous phase liquid.

Materials and method

Emulsification microchannel system

An emulsification microchannel system was assembled in-house. A transparent flow channel was used with dimensions of 2.0 mm in diameter and 0.20 m in length. A blunt tip syringe needle of 0.35 mm (ID) was used and it was inserted into the flow channel at varying angles (0 to 150 degree) relative to the continuous flow and it was carefully aligned to the center of the flow channel. A steady, dampened continuous flow of the palm oil (Buruh Brand, Malaysia) was generated in the channel with a flow rate between 2 - 300 ml/min. *k*-carrageenan (Tacara, Malaysia) of 1.5% w/w solution was delivered to the flow channel through a syringe needle with a flow rate of 0.0020 - 50 ml/min. A filtered light source was fixed behind the flow channel to enable capturing images by using a digital camera for image analysis. The system was set-up at a vibration-free table and the continuous phase was allowed to flow in the direction of the gravity.

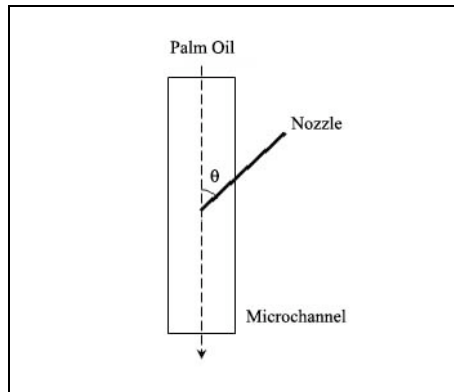


Fig 1: Schematic diagram of nozzle insertion angle

Droplet break-up behavior

The effect of flow velocity of liquids and nozzle insertion angle on the transition points between dripping and jetting modes were investigated. The velocities of dispersed and continuous phase were increased gradually and in stepwise manner in order to observe the transition points from dripping to jetting. This method was practiced for different nozzle angles.

Droplet size & size distribution

Images of *k*-Carrageenan droplets were captured by using a digital camera. Mean droplet size and size distribution were analyzed by using an image analyzer software.

Results and discussion

Effect of flow dynamics and disperse phase insertion angle on droplet breakup mode

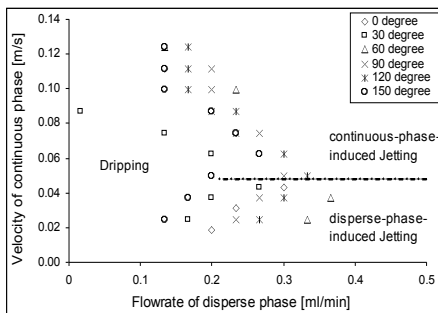


Fig 2: Droplet breakup mode at various flow combinations and injection angles

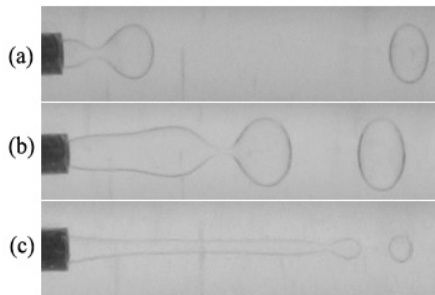


Fig 3: Three type of breakup modes: (a) Dripping, (b) Disperse-phase-induced jetting, (c) continuous-phase-induced jetting

Fig. 2 shows the effect of flow velocity of liquids and nozzle insertion angle on the transition points between dripping and jetting modes. It can be clearly seen that the break-up mechanisms of liquid droplets could be governed by three modes: dripping, disperse phase-induced jetting and continuous phase-induced jetting (refer to Fig 3), in which the transition points between these modes were affected by the liquids flow velocities and nozzle angle. In general, it was found that the dripping mode occurred at low disperse phase flow rates (i.e. $Q_{disp} < 0.1$ ml/min) where the droplets were directly formed at the nozzle outlet (Figure 3a). Increasing the flow rates caused a transition from dripping to disperse phase-induced jetting where an uninterrupted liquid jet was formed before breaking-up into droplets (Figure 3b). It was also found the dispersed phase flow rates required to cause the transition was higher with increasing continuous phase velocity ($V_{cont} < 0.05$ m/s). Interestingly, at relatively high disperse phase flow rates ($Q_{disp} > 0.3$ ml/min) and at continuous phase velocity beyond 0.05 m/s, a different type of liquid jet could be observed (continuous phase-induced jetting) (Figure 3c) where the jet was mainly caused by the flow strength of the continuous phase. The shearing effect of the continuous phase did not instantaneously cause droplet break-up but it resulted in thinning of the disperse phase liquid that formed jet stream before breaking up. In addition, the shift in the transition point between dripping and jetting was in reverse trend as the transition occurred at lower disperse phase flow rates with increasing continuous phase velocity. On the other hand, it was found that the nozzle insertion angle did not significantly change the pattern of the transition curve but it caused a shift in the curve, depending on the disperse phase flow rates.

Effect of nozzle insertion angle on mean droplet size and size distribution

Figure 2 shows the effect of nozzle insertion angle on mean size of liquid droplets formed in the dripping mode. It can be clearly seen that the droplet mean size decreased significantly with increasing angle. This could be due to the higher exposure of disperse phase liquid to the continuous phase by the increase in the nozzle insertion angle. As a result, the shearing effect could be more effective due to larger contacting area between the liquids, thus the breakup of the liquid pendant took place sooner to form smaller droplets. Figure 3 shows the effect of nozzle insertion angle on size distribution of liquid droplets formed in the dripping mode. It could be generally deduced that the nozzle insertion angle has no effect on the size distribution. This suggests that the arrangement of the nozzle did not up-set the liquid flow and droplet formation. In general, the coefficient of variance was about 5% which indicates that droplets with narrow size distribution could be produced.

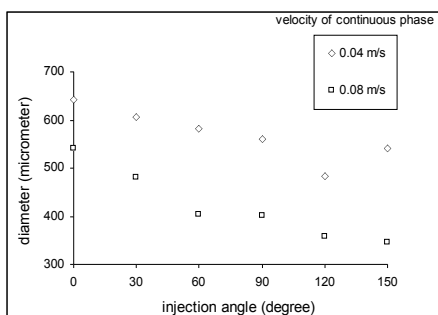


Fig 4: Effect of nozzle insertion angle on droplet size

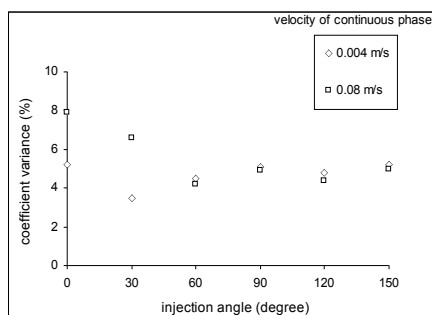


Fig 5: Effect of nozzle insertion angle on droplet size distribution

Conclusion

In conclusion, it was found that the breakup mechanisms of microchannel emulsification can be divided into 3 modes: dripping, disperse phase-induced jetting and continuous phase-induced jetting. It was found that transition points between these modes were affected by the liquids flow velocities and nozzle insertion angle. It was also found that the increasing nozzle insertion angle produced smaller droplets in the dripping mode as the effective shearing area could be higher. The nozzle insertion angle showed no significant effect on the size distribution in the dripping mode and the coefficient of variance was found to be about 5%.

Acknowledgment

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