

# IONIC LIQUID BASED DROPLET FORMATION AND MASS TRANSFER STUDY IN MICROFLUIDIC DEVICES

Lin Bai, Yuhang Fu, Shufang Zhao and Yi Cheng\*  
Department of Chemical Engineering, Tsinghua University,  
Beijing, China, 100084

## *Abstract*

Droplet formation and mass transfer performance in ionic liquids (ILs) involved microfluidic systems were studied in this work. It was found that some of the well acknowledged principles of droplet formation were still applicable in highly viscous ILs participated systems, but there are also some distinct differences on droplet size prediction and flow regime classification. In one of the most common IL-based droplet flow systems, i.e., water/IL droplet system, micro-LIF technique was applied to visualize the mass transfer performance between the two phases. Quantitative calculation of the mass transfer coefficient confirmed the enhancement effect of droplet-based microfluidics on mass transfer in IL involved systems.

## *Keywords*

Droplet-based microfluidics, Ionic liquid, Droplet formation, Mass transfer, Micro-LIF.

## **Introduction**

Ionic liquids (ILs) have attracted tremendous attention in recent years because of their superior features (low vapor pressure, high electrical conductivity, strong dissolving ability, etc.) and promising applications in chemical reaction, extraction and so on (Hallett, J.P. and Welton, T., 2011). However, one of the difficulties is that most ILs are highly viscous, leading to large mass transfer resistance. Microreactors are well known for their large surface-to-volume ratio and high mass and heat transfer efficiency. Especially, droplet based microfluidics offers not only larger surface area but also controllable operation condition. Therefore it is considered attractive to apply ILs in droplet based microreactors to enhance the mass transfer and promote the overall reaction or extraction efficiency. Previous researches have revealed that fluid viscosity could affect droplet formation involving droplet size and flow pattern distribution (De Menech et al., 2008; Gupta et al., 2009; Liu and Zhang, 2009). It is of great interest to comprehensively understand the droplet-based microfluidic performance when ILs and other high viscosity fluids are involved. Then, to test whether the droplet based

microfluidics is effective in enhancing the mass transfer in IL involved system, it is necessary to investigate the mass transfer in IL and droplet based systems. Research on mass transfer of ILs involved system is scarce, especially in the case of micro-structured reactors. Therefore we aim to investigate the mass transfer in water/IL droplet flow systems using the micro-LIF (laser-induced fluorescence) technique. This technique allows for qualitative visualization of mass transfer process and quantitative calculation of mass transfer coefficient, providing a full-scale understanding and evaluation of the mass transfer performance.

## **Experimental**

The schematic diagram of the experimental setup is shown in Figure 1. In droplet formation experiment, two different ILs, i.e., [C<sub>4</sub>mim][PF<sub>6</sub>] and [C<sub>4</sub>mim][BF<sub>4</sub>] were chosen, together with some other high viscosity fluids, such as glycerol and silicon oil. A CCD camera was used to record the droplet flow process. In the micro-LIF

---

\* Corresponding author, Dr. Yi Cheng, Tel: 86-10-62794468; Fax: 86-10-62772051; Email: yicheng@tsinghua.edu.cn.

experiment, rhodamine 6G was dissolved in water droplet which would emit fluorescence light after being illuminated by laser source. Dynamic concentration inside droplet could then be obtained, together with the mass transfer behavior.

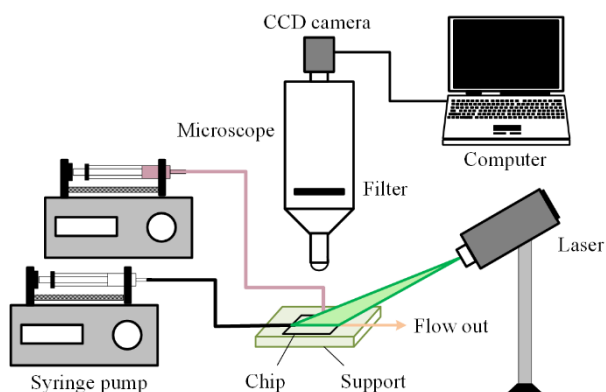


Figure 1. Diagram of the experimental setup.

## Results and discussion

For droplet flow systems incorporating IL and other high viscosity fluids, flow regime demarcation and droplet size scaling laws were systematically analyzed and compared with those in regular low viscosity fluid systems. According to our experimental results, some of the well acknowledged principles on droplet formation are still suitable in highly viscous fluid systems, but there are also some distinct differences. First, it was considered before that  $Ca$  could be adopted as criteria to discriminate between different flow regimes. However, our results showed that flow regime distribution based on  $Ca$  can be significantly different when fluid viscosities vary much. So there is hardly a unified critical  $Ca$  values that could be used to distinguish between different flow regimes. Second, it is more reasonable to use the absolute viscosity value of the continuous phase  $\mu_c$  rather than the viscosity ratio  $\lambda$  to reflect the influence of fluid viscosity on droplet size. In addition, unlike the cases of low viscosity fluid systems where droplet size could be correlated based on  $Ca$  exponentially, the effect of  $\mu_c$  becomes so significant that  $Ca$  is not enough when continuous fluids with large viscosity are applied (shown in Figure 2). Instead, a new parameter incorporating  $\mu_c$  and  $Ca$  was defined to correlate droplet size of all systems in a power law relationship.

On the base of the above droplet formation study, we further investigated the mass transfer performance in one of the IL-based droplet flow systems, i.e. water/IL microdroplet system. Thanks to the micro-LIF technique, mass transfer performance was visualized vividly. The mass transfer coefficient  $k_L a$  changing with flow velocity is shown in Figure 3. It can be seen that  $k_L a$  in this droplet flow system is in the range of  $0.05 \sim 1.5 \text{ s}^{-1}$ , which is about  $10^3 \sim 10^4$  times higher than that in conventional macro-system. This confirmed that droplet-based microsystems could indeed enhance the mass transfer between IL and water.

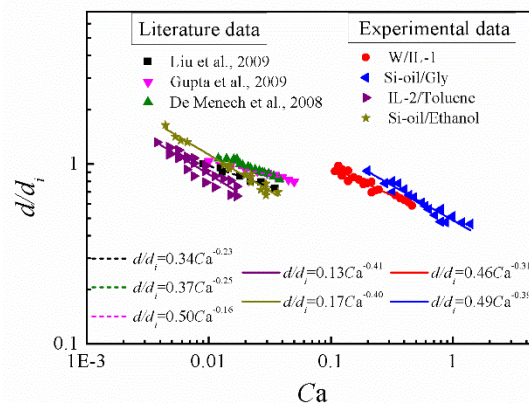


Figure 2. Droplet size in different fluid systems.

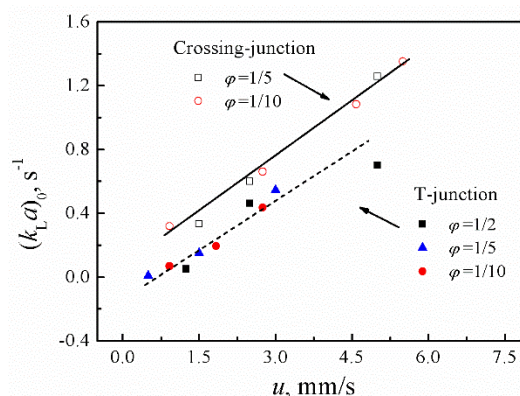


Figure 3. Change of mass transfer coefficient with flow velocity in water/IL microdroplet system ( $\phi = Q_d/Q_c$ ).

## Conclusion

The above study confirmed that ILs could be applied in droplet based microreactor, either as the dispersed or continuous phase. The mass transfer study verified that droplet-based microreactor is effective in alleviating the mass transfer problem of IL involved systems.

## Acknowledgments

Financial supports from National 973 Project of PR China (No. 2013CB733604) and National Natural Science Foundation (No. 21576151) are acknowledged.

## References

- De Menech, M., Garstecki, P., Jousse, F., Stone, H.A., 2008. Transition from squeezing to dripping in a microfluidic T-shaped junction. *J. Fluid Mech.* 595, 141-161.
- Gupta, A., Murshed, S.M.S., Kumar, R., 2009. Droplet formation and stability of flows in a microfluidic T-junction. *Appl. Phys. Lett.* 94 (16), 164107.
- Liu, H., Zhang, Y., 2009. Droplet formation in a T-shaped microfluidic junction. *J. Appl. Phys.* 106 (3), 034906.
- Hallett, J.P., Welton, T., 2011. Room-temperature ionic liquids: solvents for synthesis and catalysis. 2. *Chem. Rev.* 111 (5), 3508-3576.