

論文の内容の要旨

論文題目 Development of a Novel Microfluidic Device
by Utilizing Micro Droplet Collision and
Its Application to Chemical Processes
(マイクロ液滴衝突を用いた新規マイクロ流体デバイスの
開発と化学プロセスへの展開)

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Abstract

Microfluidic devices have achieved rapid and integrated chemical analysis by utilizing the characteristics of micro space. In this thesis, the liquid-liquid system, termed a micro droplet collider, to utilize both of the spatial and temporal localized energy simultaneously is newly introduced by adding temporal localization to microfluidics. Liquid droplets in the gas phase confined in microchannels are used for spatial-temporal localization. A highly accelerated droplet generates the high kinetic energy in the spatial localization. Rapid collision enables the spatial and temporal localization of the high-energy, which induces the conversion and transfer of the high-energy to other work in the target.

In chapter 2, a micro droplet collider device which has launchers for the droplet shot, tracks for the droplet run and a chamber for the collision was designed based on the gas-liquid Laplace pressure and fabricated on a glass substrate. The velocity of droplet (0.25 nL) reached 920 mm s^{-1} with the degree of acceleration of $2,300 \text{ m s}^{-2}$ (230 G). The droplet velocity in the micro droplet collider is 10^2 times faster and the kinetic energy of the droplet is 10^4 times higher than the conventional droplet-based microfluidic methods. Rapid ($< 1 \text{ ms}$), inelastic and minimally deformable collision between droplets was achieved, which enabled the utilization of the localized high-energy efficiently.

In chapter 3, droplet motion in acceleration and droplet internal flow in collision were investigated to clarify the basic physical properties by using a high-speed camera under microscope observation. It was confirmed that the droplet velocity reached maximum just after shot and became constant in the downstream of the microchannel track. The droplet terminal velocity increased as increasing the applied air pressure and reached 1.4 m s^{-1} when applying 400 kPa of air pressure. The fluorescent particles were seeded in the droplet to visualize the droplet internal flow and captured by a high-speed camera equipped with an image intensifier unit. The accelerated droplet was found to be penetrated into the stopping target droplet. The flow induced by collision was two orders of magnitude faster than that induced by shear stress of air flow after collision.

In chapter 4, two kinds of micro chemical processes which have not been achieved in the conventional microfluidic formats were realized by utilizing the spatial-temporal localized energy of the droplet. Rapid mixing between the nL order of droplets having a 1:10 volume ratio was achieved by inducing the internal flow by the droplet collision, which was 6,000 times faster than molecular diffusion. Integration of droplet process and parallel flow processes was achieved by injecting the accelerated droplet to the flow beyond the dynamic pressure.

The developed micro droplet collider is expected to contribute greatly to microfluidics and chemical processes on a microchip.

1. Introduction

In recent years, there has been great interest in microfluidic devices for miniaturizing chemical systems and integrating various chemical processes¹. These devices have many advantages based on spatial localization of the liquid system, including short analysis time, low consumption of sample and reagent amounts, small waste volumes, effective reaction due to the large specific interfacial area, and small space requirement. In general, microfluidics can be divided into two categories. One is the continuous (including parallel) flow format² and the other is the droplet-based format³. In the continuous flow format, for example, our group demonstrated the integration of fundamental chemical operations such as mixing, reactions, solvent extraction and multi-phase flow networking by applying surface chemistry. In the second category, the droplet-based format has focused on creating discrete volumes with the use of an immiscible phase. Primarily, water droplet-in-oil format have been widely studied. Droplet generation and manipulation such as transporting, fusion, mixing and sorting have been achieved by the steady laminar flow of the immiscible phase. All of the conventional approaches in microfluidics have intended to realize chemical processes inside the steady liquid flow and by molecular diffusion.

In this thesis, the liquid-liquid system, termed a micro droplet collider, to utilize both of the spatial and temporal localized energy simultaneously is newly introduced by adding temporal localization to microfluidics. Liquid droplets in the gas phase confined in microchannels are used for spatial-temporal localization. A highly accelerated droplet generates the high kinetic energy in the spatial localization and rapid collision enables the spatial and temporal localization of the high-energy, which induces the conversion and transfer of the high-energy to realize various novel chemical processes.

The objective of this study is to realize the micro droplet collider and to develop chemical processes. Specifically, this thesis addresses the following points. (1) The micro droplet collider is realized on a microchip (2) Dynamics of the micro droplet collider is investigated. (3) Chemical processes by utilizing the spatial-temporal localized liquid energy are developed.

2. Realization of a Micro Droplet Collider

In this chapter, the chip design to realize a micro droplet collider and performance evaluation of droplet acceleration and collision are described.

2.1 Chip design

The designed microchip (Fig. 1) has the launcher for droplet shot, the microchannel track for droplet run, and the collision chamber for droplet collision. Through the whole chip design, the gas-liquid Laplace pressure is utilized. In the launcher, the droplet can be generated at the shallow hydrophobic Laplace valve and be accelerated by applying air pressure (Fig. 2). In the chamber, droplet collision can be conducted by holding the target droplet by Laplace pressure and eliminating air to the shallow hydrophobic side channel (Fig. 3).

2.2 Performance evaluation

The velocity of droplet (0.25 nL) reached 920 mm s^{-1} with the degree of acceleration of $2,300 \text{ m s}^{-2}$ (230 G). The droplet velocity in the micro droplet collider is 10^2 times faster and the kinetic energy of the droplet is 10^4 times higher than the conventional droplet-based microfluidic methods. The rapid ($< 1 \text{ ms}$), inelastic collision between droplets with minimally deformation was achieved in the micro droplet collider, which enabled the efficient utilization of the spatial-temporal localization energy in the liquid phase.

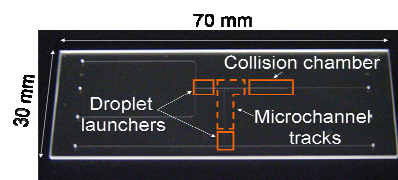


Fig. 1. Micro droplet collider chip.

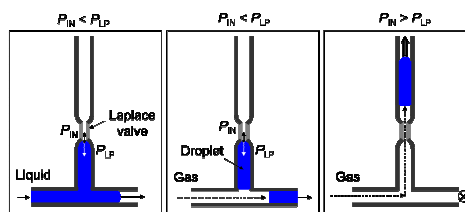


Fig. 2. Droplet shot from the launcher.

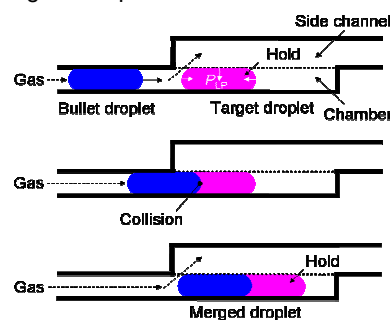


Fig. 3. Droplet collision in the chamber.

3. Investigation of Dynamics of the Micro Droplet Collider

In this chapter, dynamics of the micro droplet collider is investigated to optimize the system, to realize extreme performances, and to develop novel micro chemical processes.

3.1 Investigation of a Droplet Motion in acceleration

The 1.7 nL of a droplet was generated and shot from the launcher by 20 kPa of air pressure. The droplet motion was captured by a high-speed camera (1,000 ~ 10,000 fps) under a microscope observation. It was confirmed that the droplet velocity reached maximum just after passing the shallow Laplace valve part and became constant in the downstream of the microchannel track as shown in Fig. 4. The droplet terminal velocity increased as increasing the applied air pressure and reached 1.4 m s^{-1} when applying 400 kPa of air pressure as shown in Fig. 5. Modeling of droplet motion and determination of dominant resistance force are described in this thesis.

3.2. Investigation of a Droplet Internal Flow in Collision

Droplet internal flow in the collision event was investigated to estimate how the energy of the accelerated droplet transfers and induces flow to the target droplet. The fluorescent particles were seeded in the droplet to visualize the flow and captured by a high-speed camera equipped with an image intensifier unit. Droplet internal flow induced by collision is shown in Fig. 6. The accelerated droplet was found to be penetrated into the stopping target droplet. The flow induced by collision was two orders of magnitude faster than that induced by shear stress of air flow after collision.

The results obtained in this chapter are useful to apply to chemical processes.

4. Application to Chemical Processes

In this chapter, micro chemical processes that have not been achieved yet are realized by utilizing the spatial-temporal localized energy of the droplet.

4.1. Mixing between Droplets Having a Large Volume Ratio

Mixing is basic and important in various bio/chemical processes. Mixing between droplets, especially in a large volume ratio, on a microscale is challenging because of a laminar flow regime and time-consuming molecular diffusion process. However, rapid mixing of two droplets having a 10:1 (Fig. 7) and 1:10 (Fig. 8) volume ratio was achieved by the collision of the accelerated droplet to the target. Mixing time was estimated as 0.5 s, which is more than 6,000 times faster than that of molecular diffusion.

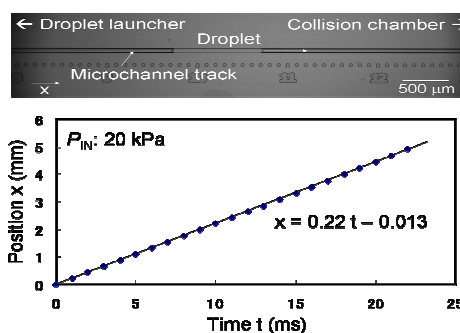


Fig. 4. Droplet behaviors in the track. (upper) Captured image. (lower) Analyzed droplet position against time.

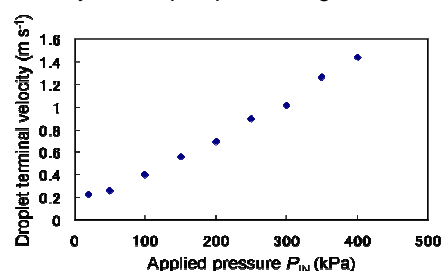


Fig. 5. Droplet terminal velocity against the applied air pressure.

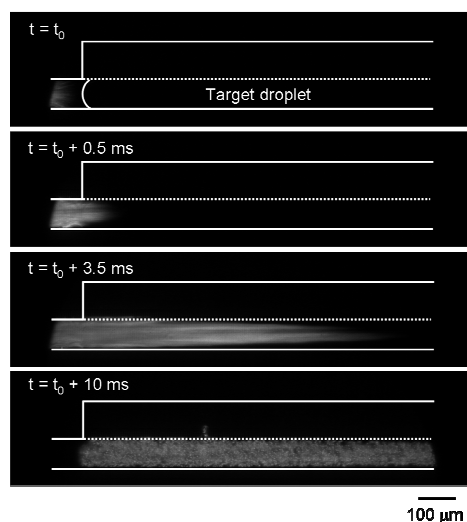


Fig. 6. Droplet internal flow induced by collision.

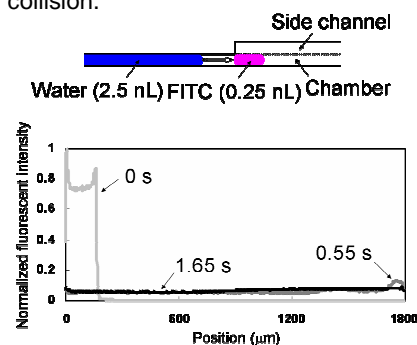


Fig. 7. Results of mixing between droplets having a 10:1 volume ratio.

4.2. Integration of Droplet and Parallel Flow Processes

Complicated chemical processes have been achieved under a network of parallel flow. In parallel flow processes, however, the batch processes such as liquid volume metering and injection of the metered liquid to the flow are difficult. Injection of a droplet to the flow has been difficult in the conventional droplet-in-oil format, because of the dynamic pressure of the flow and the difficulty of the oil phase elimination. Here, microchip was designed to control both the parallel flow and the droplet independently by utilizing the gas-liquid Laplace pressure. The droplet was accelerated toward the parallel flow and successfully injected beyond the dynamic pressure as shown in Fig. 9.

The developed micro chemical processes in this chapter would contribute to the bio/chemical analysis and extend applications of the microfluidic devices.

5. Concluding Remarks

In this thesis, the novel microfluidic device, termed the micro droplet collider, was developed. Micro chemical processes which have not been achieved yet were realized by the collision of the accelerated droplet to the target in the gas phase. In the future, bio/chemical analysis and reaction will be realized by utilizing the developed device.

References

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Journal Publications

- (1) K. Takahashi, K. Mawatari, Y. Sugii, A. Hibara, T. Kitamori, "Development of a micro droplet collider; the liquid-liquid system utilizing the spatial-temporal localized energy", *Microfluidics and Nanofluidics*, in press.
- (2) K. Takahashi, Y. Sugii, K. Mawatari, T. Kitamori, "Investigation of the droplet acceleration and collision in the gas phase in the micro droplet collider", *Microfluidics and Nanofluidics*, in preparation.
- (3) K. Takahashi, Y. Sugii, K. Mawatari, T. Kitamori, "Investigation of dynamics of the spatial-temporal localized liquid-liquid system", *Physical Review Letters*, in preparation.
- (4) K. Takahashi, K. Mawatari, Y. Sugii, T. Kitamori, "Integration of the droplet processes and continuous flow processes by the micro droplet collider", *Lab on a Chip*, in preparation.

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- (1) K. Takahashi, K. Mawatari, A. Aota, A. Hibara, T. Kitamori, "Micro droplet collider: its application to micromixer", *Proc. Micro Total Analysis Systems 2009*, vol. 2, 1413-1415 (2009).
- (2) K. Takahashi, Y. Sugii, K. Mawatari, T. Kitamori, "Dynamics of a micro droplet collider to extend microfluidic applications", *Proc. Micro Total Analysis Systems 2010*, submitted.

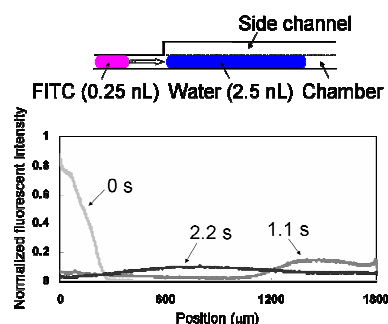


Fig. 8. Results of mixing between droplets having a 1:10 volume ratio.

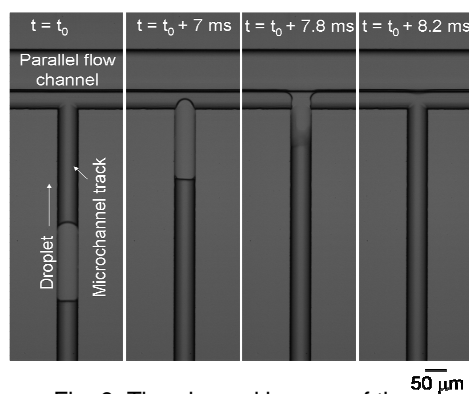


Fig. 9. Time-lapsed images of the droplet injection to the parallel flow