Microchannel Reactor (Stacked Multi-Channel Reactor: SMCR®) for Bulk Chemical Industry

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A microchannel reactor (MCR) has high thermal performance and rapid mass transfer thanks to its small channels where the reaction takes place. The microchannels offer a field that is more reactive than that offered by conventional mixer-type reactors. The application of MCRs, however, is limited to products such as medicines that have high value and are produced in small volumes. This is due to the limitation of the flow capacity of an MCR. Against this background, Kobe Steel has developed a stacked multi-channel reactor (SMCR®) that enables mass production and is applicable to the bulk chemical industry. This report explains the technology of the MCR, the features and the construction of the SMCR and the use of an SMCR for development work aimed at commercialization.

Introduction

Microchannel reactors (MCRs) are receiving considerable attention. They have flow passages with small diameters, which enables them to achieve a heat transfer performance and mass transfer rate higher than those achieved by conventional reactors such as mixing tanks. These MCRs, however, have been applied only in the small-volume production of high-value-added products, such as pharmaceuticals that are manufactured in a wide variety. This is due, among other things, to the limitation in their throughput capacities.

Against this background, Kobe Steel has developed a stacked-type multi-channel reactor (hereinafter an SMCR®, note 1)) capable of high volume processing and has been promoting the applications of this new MCR technology to bulk chemicals.

This paper introduces the technologies of MCRs, the structure of the SMCR and its features. Also introduced is the flow of the development of the SMCR up to its commercial use.

1. What is an MCR?

1.1 Features of MCR

Many have reported that reducing the diameters of flow passages, as shown in **Fig. 1**, achieves a higher heat transfer performance and mass transfer rate compared with conventional reactors such as mixing tanks.1), 2)

Fluid in a small flow passage is greatly affected by wall surfaces, but is relatively less affected by gravity. This makes the separation commonly observed in conventional equipment and piping–that is, a separation between heavy and light products, or between gas and liquid–less likely to occur. As a result, various flow patterns appear, as shown in **Fig. 2**, depending on the physical properties of the fluid and the flow velocity. In the case of two mutually insoluble liquids such as water and oil, for example, slug flow appears in the region where the flow velocity is relatively low, while two-layer flow appears in the region where the flow velocity is high. **Fig. 3** shows a photograph of slug flow with its image diagram. When the wall

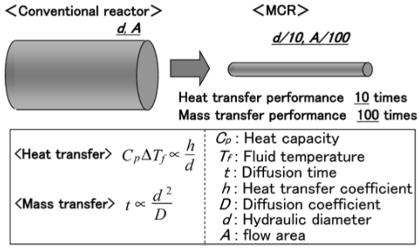

Fig. 1 Comparison between conventional reactor and microchannel reactor

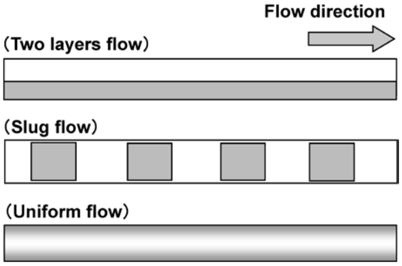

Fig. 2 Example of flow pattern in microchannel

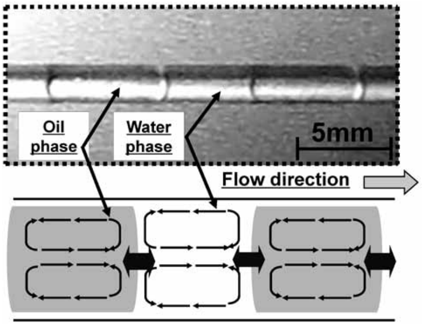

Fig. 3 Slug flow in microchannel

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note 1) SMRC is a registered trademark of Kobe Steel.

surface is either stainless steel or glass, for example, its hydrophilic nature makes the water stagnate on the wall surface. Thus the oil flow is enveloped by the water. As a result, the slug flow sometimes creates a greater contact area at the interface of the fluids than does the two-layer flow. In addition, such wall surfaces also have been confirmed to cause an internal circulation flow.[3), 4)] Hence, a higher mass transfer performance can be achieved by selectively exploiting the slug flow, which is expected to further improve the performance of MCRs.

1.2 Problems with conventional MCR

An MCR is often called a "micro reactor" because of the established image of the equipment itself as being small in size, and not just because of the size of its flow passages. For this reason, the units are mostly used for small volume production, from several to several hundreds of kilograms per annum, of high-value-added products such as pharmaceuticals. They have hitherto not have been adopted by the general chemical industry, which processes large volumes amounting from several thousands to several tens of thousands of tonnes per annum.

The major reason is the high cost of making equipment, including the cost of fabricating the flow passages, which only permits the making of small units. There is also a methodological difficulty in fabricating a large number of flow passages in a unit (hereinafter referred to as "numbering-up"), making the unit unsuitable to high-capacity applications.

In short, the major issue is that, notwithstanding the reactor's potential for delivering a better performance than has ever been seen before, no means has been available for the industrial use of the MCR. It is against this background that Kobe Steel launched its development of an MCR unit capable of processing large volumes.

2. Structure of SMCR equipment

2.1 Basic technology of stacked heat exchanger

The basic technology of the SMCR lies in its brazed aluminum plate-fin heat exchangers (hereinafter called ALEX®[, note 2)]), a high-performance heat exchanger made of aluminum alloy having excellent heat transfer performance. It was developed as a heat exchanger for equipment that separates air into oxygen and nitrogen at very low-temperatures. Being capable of exchanging heat among several fluids at a time, the ALEX has recently been widely used in plants as an apparatus for natural gas processing and in cryogenic section in ethylene plants.

As shown in **Fig. 4**, an ALEX comprises a brazed core for exchanging heat and a header/nozzle for guiding fluid into the core. It has a stacked structure, made capable of homogeneously distributing not only gas and liquid, but also two-phase flow (gas-liquid mixed phase), by arranging and combining fins in each layer. The apparatus can also exchange heat among several fluids simultaneously.

In some applications, an ALEX can have a heat transfer area per unit volume of 1,000m^2/m^3 or greater, which exceeds that of conventional shell & tube type heat exchangers by a factor of 5 or more; so it can enable the downsizing of the equipment. The ALEX can be used as a single unit, in which multiple cores are welded together and attached to a common header and nozzle, or as multiple units connected by pipes. This enables the ALEX to handle any given flow rate and to process large volumes.

2.2 Technologies applicable to large capacity SMCRs

Being widely used as a heat exchanger, the ALEX offers the following basic technologies:

(1) Production technology including joining and flow passage fabrication.
(2) Heat transfer design used for performance calculation, and technology for pressure loss calculation.
(3) Technologies for homogeneously distributing and mixing on the basis of the structure for gas-liquid distribution.
(4) Technology for reducing maldistribution of

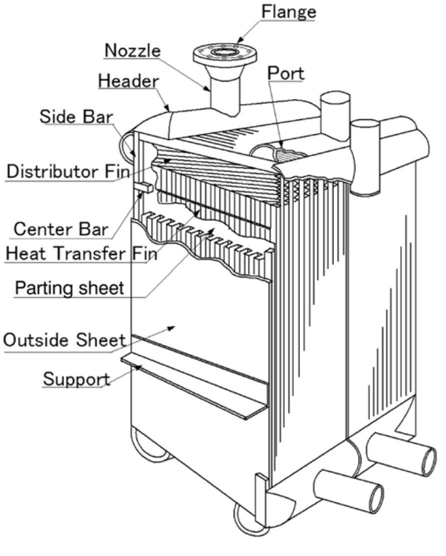

Fig. 4 Structure of ALEX

[note 2)] ALEX is a registered trademark of Kobe Steel.

the fluid between cores.

(5) Technology for reducing maldistribution of the fluid between layers.

These are the design technologies and know-how obtained through experience with the ALEX, and they can be exploited for large capacity SMCRs.

2.3 Basic construction of SMCR

The flow passages in an MCR basically consists of the combination of multiple tubes, each having a Y shape or a T shape, as shown in **Fig. 5** top. However, if used as-is for the numbering-up of capacity enlargement, this configuration is limited in the arrangement of flow passages due to difficulties in, for example, the supply method of fluids. Notwithstanding the ease of numbering-up in the stacking direction, it is difficult to arrange multiple fluid passages in an efficient manner in the width direction, rendering the structure unsuitable for large capacity MCRs. Therefore, a structure has been devised, on the basis of the existing structure of ALEX (Fig. 5 bottom), with the flow passages fabricated in a three-dimensional arrangement on both sides of each plate.

This structure allows for a dense arrangement of the flow passages within a plate, as shown in **Fig. 6**. This significantly increases the number of flow passages per unit volume, enabling high-volume processing. For increased processing volume, a plurality of plates can be stacked, as shown in **Fig. 7**, to further increase the number of flow passages. In other words, the number of flow passages per unit is given by the number of flow passages per plate multiplied by the number of the plates stacked.

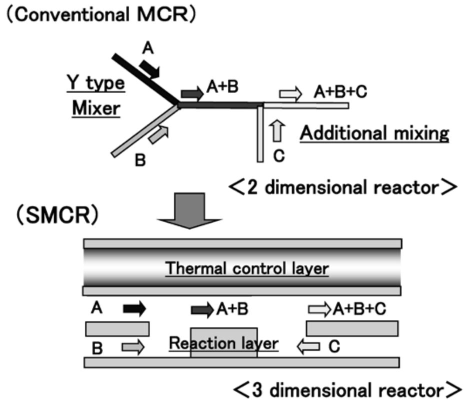

Fig. 5 Basic construction of two-dimensional reactor and three-dimensional reactor

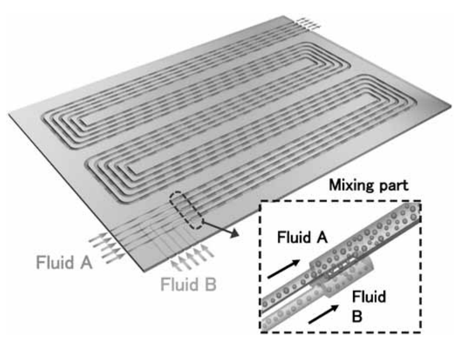

Fig. 6 Basic construction of multi-channel SMCR

When temperature control is required for operation, a thermal control layer can be stacked, as shown in Fig. 5, to permit precision temperature adjustment. For the supply of fluid into each flow passage, a header-nozzle assembly, as shown in Fig. 4 for the case of the ALEX, may be used for the homogeneous distribution of fluid into each plate (Fig. 7). A large capacity MCR, adopting the above construction, is referred to as an SMCR.

The process of making an SMCR is as follows: First, a flow passage pattern as shown in **Fig. 8** is formed by, for example, chemical etching on metal plates, such as stainless steel plates. Next, these plates are stacked, in combination with thermal control layers, until the required number of flow passages is reached. This is followed by heating and pressing the stacked plates in a vacuum furnace to diffusion-bond them so as to form the flow passages. **Fig. 9** shows an example of a diffusion-bonded section of stainless steel. No deformation of the flow passages was found, and the crystal grains were observed to have grown beyond the bonded interface, ensuring a bonding strength equal to or higher than the strength of the base material. Hence, the pressure-withstanding performance can be estimated by a strength calculation based solely on the size of the flow passages. In addition,

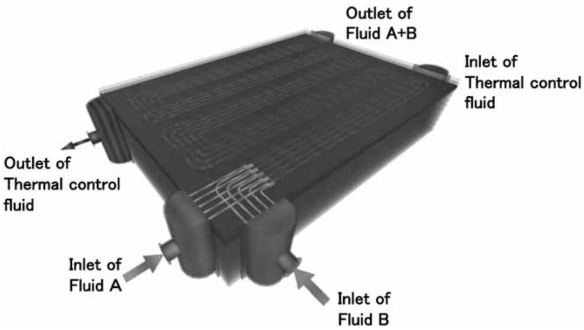

Fig. 7 Inside image of SMCR

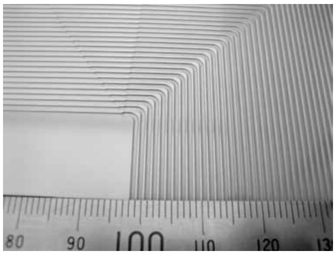

Fig. 8 Microchannel manufactured by chemical etching

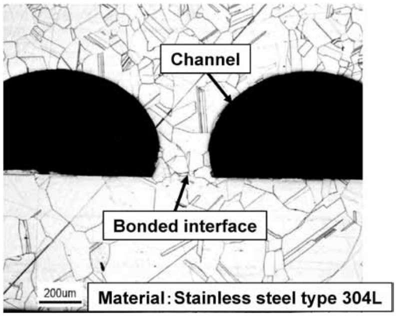

Fig. 9 Cross-sectional observation of channels and diffusion bonded interface

various materials can be used, depending on the specification requirements, such as high resistance to corrosion and being able to withstand high temperatures; this permits a high degree of freedom in design and production.

3. Flow of development up to commercial use

As in the case of conventional equipment, the development of an SMCR follows the flow shown in **Fig.10**. Here, the concept of numbering-up, a feature of the MCR, can be adapted to the SMCR so as to shorten the development period.

The following describes an example of laboratory testing. An extraction feed of dodecane containing 0.1wt% of phenol was prepared. This solution was subjected to the extraction of phenol using water as the extraction solvent. Various flow passage diameters and shapes (semicircular and circular) were used. As shown in **Fig.11**, the volumetric mass transfer coefficient, *Ka*, an indicator of the mass transfer rate, has been confirmed to correlate with the hydraulic equivalent diameter (= four times the cross-sectional area of the flow passages divided by the length of wetted perimeter).[5)]

This result verifies that laboratory test data obtained by universities and corporate research institutions can be adopted as-is for the SMCR, minimizing the overlap of testing and shortening its test and development period.

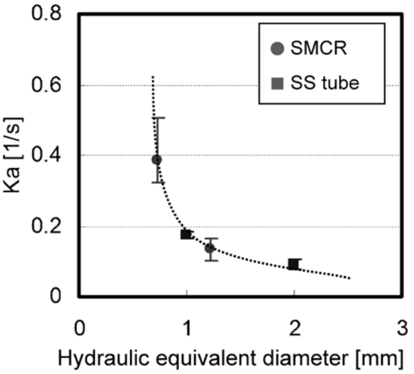

Fig.11 Relationship between *Ka* and hydraulic equivalent diameter

Bench tests are effective in predicting the performance of the commercially used equipment. These bench tests are conducted after determining the shape, plate size and the number of stacks required for commercially used equipment. Plates that have a size equivalent to that of the actual equipment, as shown in Fig. 6, are stacked in three layers, for example, or in any number of layers that allows confirmation of the effect of maldistribution. The issues involved in commercialization include the homogeneous distribution of fluid into each plate layer at the time of upsizing. In the case of ALEX, more than a hundred layers were stacked while distributing the fluid homogeneously into each flow passage. This proven design technology can be used to prevent any problems due to maldistribution in practice. Hence, the risk of performance decrement caused by scale-up, a problem for the reactors of mixing-tank type, can be significantly reduced. In the case of the SMCR, a verification test serving as both a bench test and demonstration plant is considered sufficient for making all the judgments needed for commercialization.

As described above, SMCRs are not only excellent in heat transfer performance and mass transfer rate, but they also make it possible to decrease the amount of investment and shorten the development period.

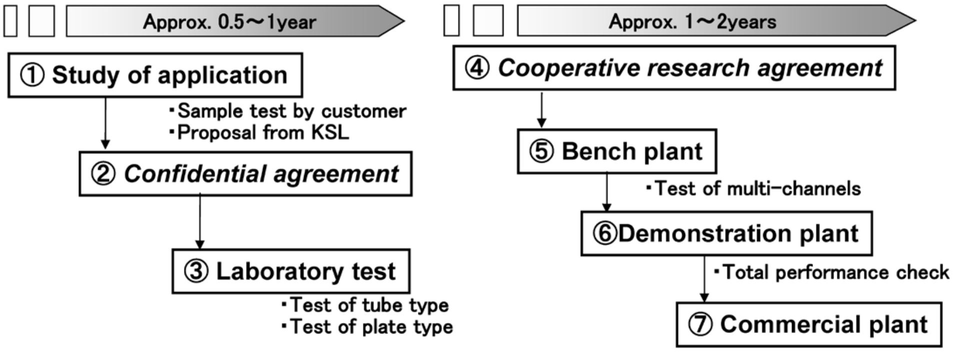

Fig.10 Flow of development work for commercialization

4. Examples of SMCR applications

4.1 Study for extraction applications

A process of making various chemical products involves an extraction step, using extraction solvent for removing objective substances (or, non-objective substances) in an objective fluid. When the extraction solvent is recycled, for example, a series of processes are performed, including extracting product from the objective fluid using extraction solvent in a mixing tank, separating the product from the extraction solvent using the difference in their specific gravities, and subsequently separating and recovering the subjective fluid and extraction solvent by a distillation operation or the like. In this case, the capacity of the solvent recovery apparatus is determined so as to match the capacity of the mixing tank used for the extraction.

When used for such extraction equipment, the SMCR is expected to provide the following effects:

(1) decreased extraction time
(2) decreased size of the extraction process equipment
(3) reduced unit consumption, thanks to using a smaller amount of extraction solvent
(4) capability of continuous processing when multi-stage extraction is required

The following describes an extraction test using an SMCR to confirm its applicability to the extraction process.

4.2 Experimental method and results

SMCR test units comprised a stainless steel plate having semicircular microchannels formed by etching, in which the stainless steel plate was sandwiched by other stainless steel plates. Three types of units having different number of flow passages were prepared: one having one flow passage (1 passage/layer × 1 layer), another having five flow passages (5 passages/layer × 1 layer), and yet another having 25 flow passages (5 passages/layer × 5 layers). These units were tested to confirm if there were any performance decrement due to maldistribution of fluid caused by the increased number of flow passages, or by the capacity upsizing. Extraction experiments were carried out using the bench test apparatus shown in **Fig.12**.

In this experiment, phenol was extracted from an extraction feed of dodecane containing 0.1wt% of phenol, using water as the extraction solvent.

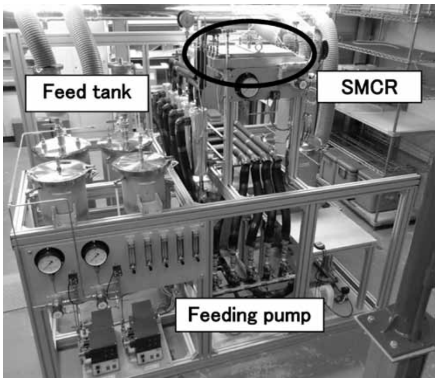

Fig.12 Bench test apparatus of SMCR for extraction use

The above feed and solvent, mixed at a volume ratio of 1, were pumped into each test unit at a given flow rate (total of 1 to 10ml/min per flow passage). The recovered solution was separated into an organic phase and a water phase. The separated organic phase was subjected to absorption spectrophotometry to measure the phenol concentration in it and to determine the extraction rate of the phenol.

A mixer extraction test was conducted, in which the extraction feed and extraction solvent, 100ml of each, were poured into a 200ml beaker and the extraction solvent phase was stirred at a given rpm using a magnetic stirrer. The phenol concentration in the extraction feed was analyzed at a given time interval to determine the extraction rate. **Fig.13** shows the experimental results. The vertical axis represents the equilibrium extraction ratio (= extraction rate (%) / equilibrium extraction rate (%)), and the horizontal axis represents the retention time.

In this mixer extraction test, increased revolution of the stirrer shortens the time required for extraction; however, revolutions higher than 400rpm resulted in a dispersion of the extraction feed in the extraction solvent, making separation difficult. It took approximately 100minutes for the mixer extraction to reach an equilibrium extraction, while SMCR achieved the same result within 0.1 to 1 minute, a shortening of approximately 1/100. The fluid from the SMCR test unit was separated into the two phases of extraction feed and extraction solvent as soon as the fluid was discharged. This is because

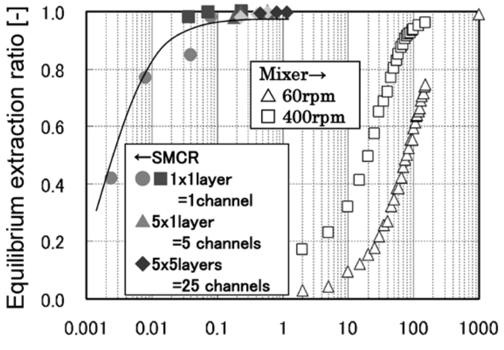

Fig.13 Test results for extraction use (SMCR vs. Mixer)

the mixing portions, shown in Fig. 5, do not vigorously mix but rather form a slug flow and/or a two-layer flow of oil and water so as to keep them separable. These experimental results confirm the following advantages of using the SMCR for extraction:

(1) The retention time is shortened to 1/100 of that required for mixer extraction.
(2) Excellent separability is achieved after the extraction.
(3) Excellent distribution of the solution is achieved regardless of the numbers of flow passages and layers.

4.3 Study of the commercial use of SMCR

Many extraction applications require multiple steps of extraction until a desired extraction concentration is reached. In a conventional extraction unit, the mixing tank serves also as a separation tank as shown in **Fig.14**. In such a tank, multiple steps of extraction, each followed by a separation step, were performed in batches. In some extraction applications, the extraction operation itself lasts for only several minutes; however, the separation takes several hours, requiring much time and labor to achieve the target extraction rate.

On the other hand, when the SMCR is used for multistage extraction, more than one extraction unit can be stacked and integrated as shown in **Fig.15**. Furthermore, the excellent separability after extraction enables a continuous process: the raw material, rapidly separated by settlers, is continuously fed into the SMCR.

This eliminates the time-consuming separation and the switching operation of charging and discharging the solution, steps required by the conventional-type process, thus enabling the SMCR to perform efficient extraction. Hence, there are two options for the commercial use of the SMCR. One is to minimize the equipment size with a reduced process volume per unit time so as to complete the process within a time equivalent to that required for a conventional batch process. The other is to aim at a larger process capacity by processing at higher efficiency within a time shorter than that required by a batch process.

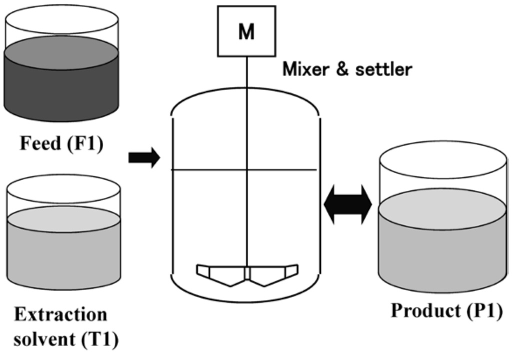

Fig.14 Conventional extraction unit using mixer

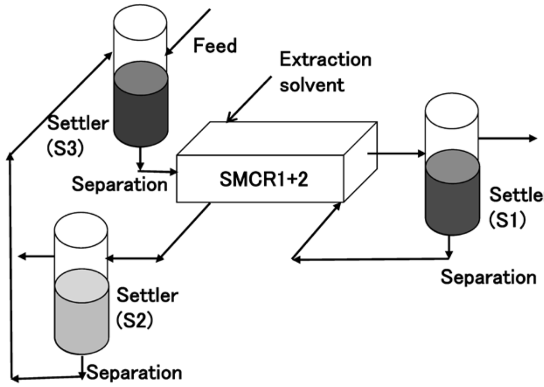

Fig.15 Multi-stage extraction unit using SMCR

As future efforts to promote commercial use, testing will be continued for other extraction applications to accumulate data on their extraction performance and to compare their economy, including equipment and operational costs, with that of a conventional extraction unit.

Conclusions

Features of MCRs such as their high heat transfer performance and mass transfer rate have led to their being considered for industrial use. In general, however, these units are small and costly, which limits their application to high-value-added products and to cases where a rapid reaction calls for almost no retention time and only small equipment is required. However, the SMCR described in this paper can have increased capacity while keeping the MCR's functions of high heat transfer and mass transfer. This makes these units applicable not only to high-value-added products, but also to bulk chemical industries involving, for example, extraction and other reactions requiring a relatively long retention time.

The SMCR has various features, including high heat transfer performance and high yield. Thus, its applications are not limited to the downsizing of equipment. Its advantage of continuous-processing capability is exploited to improve production efficiency, including the elimination of setup time that would otherwise be required if using conventional batch processes. Versatile effects, including the relaxing of process conditions (e.g., operating pressure and temperature), can be achieved. This can save energy and reduces the amount of the various chemicals used, including extraction solvent.

References

1) J. Yoshida. *Development and Application of Microreactors*. CMC Publishing CO., LTD., 2003, p.4.
2) G. S. Calabrese. *AIChE J.* 2011, Vol. 57, No. 4, p.828.
3) A. Ghaini et al. *Chemical Engineering Science.* 2011, Vol.66, p.1168-1178.
4) H. Kinoshita et al. *Lab Chip*, 2007, Vol. 7, p.338-346.
5) A. Matsuoka et al. Extraction performance in small flow channel with semicircular cross section. *Proceeding of the SCEJ 44th Autum Meeting 2012*, I305.