Large Capacity Reactor, Stacked Multi-Channel Reactor (SMCR™) for Flow Chemistry

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Abstract

The stacked multi-channel reactor (SMCR™) is a (continuous) flow chemical reactor that replaces the conventional stirred bed reactors and has been developed as a reactor capable of large-capacity processing. As an effort after the development, an SMCR™ made of metal has been added to the lineup and, furthermore, a ceramic SMCR™ with high corrosion resistance has been developed to expand the applications, for which verification tests have been conducted against thermal shock. Moreover, basic tests and bench scale testing equipment have been actively commissioned and implemented with the aim of promoting the commercialization of SMCR™ in addition to equipment supply. As for bench scale testing, a bench scale testing apparatus installed in Kobe Steel performs verification tests to verify the usefulness of SMCR™ units.

Introduction

Efficient reaction can be achieved by simply flowing two different fluids through a fine channel with a diameter of approximately 1 mm. This is the principle of microchannel reactors. For the chemical manufacturing process in Japan and overseas, flow synthesis (continuous synthesis) in the fine space of a microchannel reactor is attracting attention from the viewpoint of improving efficiency, quality stability, and safety.[1), 2)] The main focus has hitherto been placed on laboratory-level studies and application studies on products in small quantities with high additive value, such as those in the pharmaceutical field. However, full-scale studies are now being conducted on applications that require large-capacity processing of fine chemicals and bulk chemicals, for example.

Kobe Steel has developed a stacked multi-channel reactor (hereinafter referred to as the "SMCR™" Note 1)) as a microchannel reactor capable of large-capacity processing for flow synthesis, with the aim of applying it to commercial production plants. This paper describes the fundamental configuration and characteristics of the SMCR™, Kobe Steel's efforts toward its application to commercial production, and its future perspective.

1. Characteristics and applicable use of SMCR™

1.1 Fundamental configuration and characteristics of SMCR™

In general, a microchannel reactor comprises microchannels of sub-millimeter to several millimeters in diameter, such that fluids meet inside the apparatus. The greatest characteristic of microchannel reactors is that these micro channels serve as reaction fields to achieve high heat transfer performance and mass transfer rate.[3)] **Fig. 1** compares a conventional stirred-bed reactor

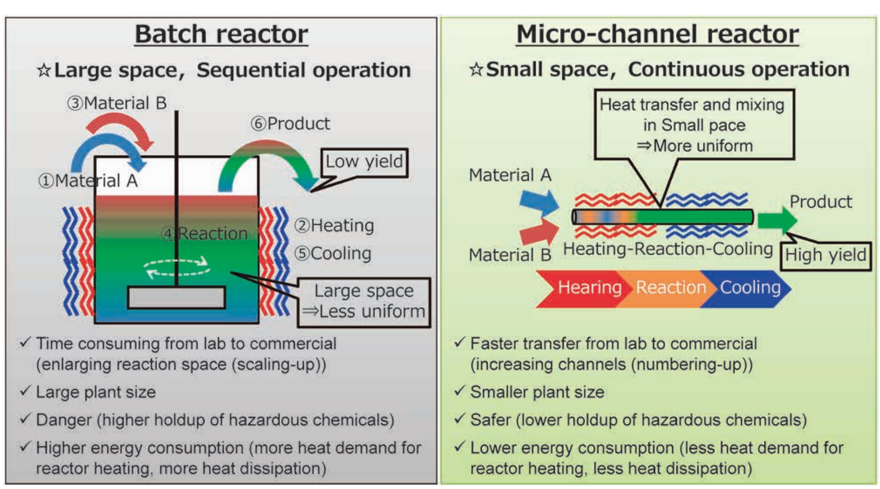

Fig. 1 Comparison between batch reactor and micro-channel reactor

Note 1) SMCR is a registered trademark of Kobe Steel, Ltd.

and a microchannel reactor. The throughput of each channel in a microchannel reactor is generally small, hence the throughput is increased by using a technique called "numbering-up," by which multiple channels are arranged.

The SMCR™ is a unique product developed by Kobe Steel. This apparatus can easily be enlarged in capacity by the numbering-up of channels using chemical etching and diffusion bonding. **Fig. 2** shows the basic configuration of the channels. Multiple channels are formed on each metal plate by chemical etching, and a plurality of such plates is stacked to be firmly bonded by diffusion bonding. With such a configuration, a reactor with 10,000 or more channels can be made. The number and length of the channels inside an SMCR™ can be freely designed depending on the reaction conditions. In addition, some of the plates may be used for heat medium channels to allow the adjustment of the reaction temperature.

With such characteristics, the SMCR™ is a reactor that can simultaneously achieve capacity enlargement, continuous processing and apparatus downsizing for various reaction applications, and there is a strong need for their commercialization. **Fig. 3** shows the overall and inside images of the SMCR™.

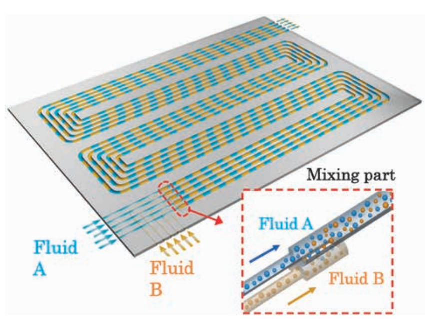

Fig. 2 Basic configuration of multi-channels in SMCR™

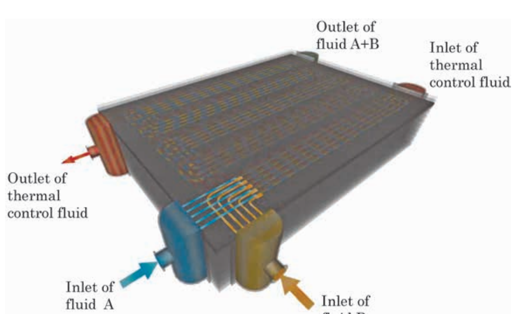

Fig. 3 Overall and inside image of SMCR™

1.2 Applicable use of SMCR™

The main applicable uses of SMCR™ are considered to be "synthesis" and "extraction" in each unit operation of the chemical manufacturing process. Various organic synthesis and polymer syntheses occur at high rates with great heat of reaction, making temperature control of the synthesis difficult when conventional stirred-bed reactors are used. The use of an SMCR™ for such synthesis enables precise temperature control and raises expectations for product quality stability and higher process efficiency.

In the extraction application, on the other hand, the use of fine channels increases the contact area, making the extraction of the target substance more efficient. In addition, since no active fluid mixing is performed in the channel inside the SMCR™, the phase separation performance after extraction is significantly improved compared with the conventional stirred-bed reactors. The expected applications that can exploit these merits include the removal of impurities and catalysts after reactions and the recovery of rare metals.

2. Toward the commercialization of SMCR™

Fig. 4 shows the main steps in development aimed at commercial production based on the SMCR™. Basically, fundamental laboratory testing is performed first to optimize the reaction conditions in a manner similar to the development of the conventional chemical process. After that, bench-scale testing is performed using a SMCR™ with multiple channels to confirm any differences from the results of the basic testing. Finally, the performance and operation control are checked

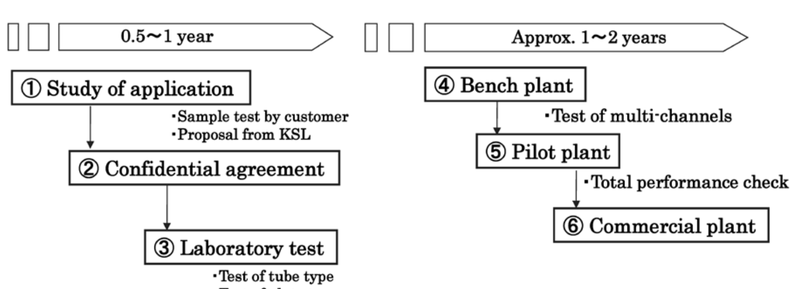

Fig. 4 Development workflow for commercialization

by pilot testing using a plant that has the same function as that used in commercial production, and the process shifts to commercial production. As mentioned earlier, when SMCR™ is used, a technique called numbering-up can be adapted to increase the throughput and to proceed to commercialization. Therefore, the reaction conditions obtained by the basic testing can be used as they are for commercial production, which reduces the risks in developing chemical processes and raises the expectation of cost reduction and period shortening.

Since the SMCR™ can be disposed in parallel or in series, there is theoretically no upper limit to capacity enlargement. In terms of configuration, it has enough functions for commercialization. On the other hand, for highly corrosive applications, metals that can be etched or diffusion bonded cannot be used. Therefore, it is necessary to solve the problems associated with the materials in order to expand the application.

Hence, Kobe Steel has been conducting the development of the ceramic SMCR™ in an effort to further promote the commercial production of the SMCR™. Furthermore, the company has been accepting consignment tests for the basic examination stage and bench stage in order to enhance the process (software) toward aiming at commercialization. The following sections introduce the details of the efforts and their results.

3. Development of ceramic SMCR™

Initially, the SMCR™ had only one lineup of apparatuses made of metals, mainly stainless steel. There are, however, many applications that cannot be handled by metallic apparatuses, e.g., applications that require corrosion resistance such as processes that use large amounts of acids and alkalis, and applications in the pharmaceutical, semiconductor and food industries, that permit no elution of metallic ions. The ceramic SMCR™ was developed with the aim of expanding its use to such applications.

3.1 Characteristics of ceramic SMCR™

Ceramics generally have high corrosion resistance and also have high strength at high temperatures. On the other hand, they are prone to brittle fracture, and caution is required during operations involving rapid heating and rapid cooling. Kobe Steel has optimized the configuration of SMCR™ by structural analysis and developed a ceramic SMCR™ with excellent thermal shock resistance. More specifically, the configuration includes a metal or resin flange separated from the ceramic core of the main body, in which fluids are supplied and extracted from the flange, as shown in **Fig. 5**. The ceramic SMCR™ has the following characteristics:

① Discontinuous shapes in the fluid supply have been eliminated from the ceramic body, leaving only straight channels. As a result, the stress concentration has been relaxed to ensure configuration with high thermal shock resistance (see **Fig. 6**).

② The channels were arranged as much as possible in the ceramic core so as to minimize the ceramic parts without channels, which are insensitive to temperature change. This has made it possible to equalize the temperature distribution in the apparatus when the fluid supply has a temperature different from that of the ceramic core, and thus to minimize the occurrence of thermal stress.

3.2 Implementation of thermal load test

In order to verify the thermal shock property, a ceramic SMCR™ with the configuration described in Section 3.1 and the specifications shown in **Table 1** was prepared to perform a thermal load test. In other words, it was verified whether or

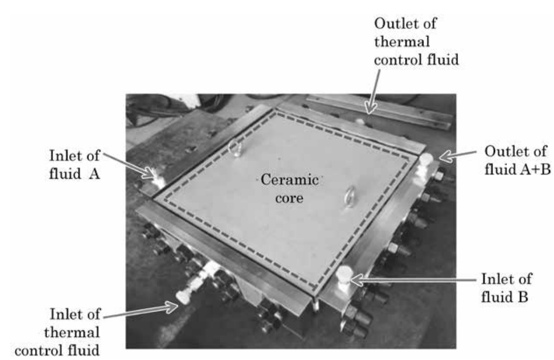

Fig. 5 Ceramic SMCR™

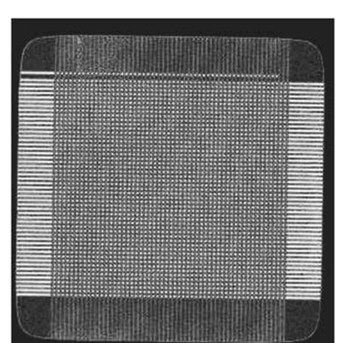

Fig. 6 Channel configuration inside ceramic SMCR™

Table 1 Specifications of ceramic SMCR™

Size (mm)	465 × 505 × 185
Weight (kg)	75
Channel specification	Size: 2 (mm) rectangle Length: 36 (m) Number: 1 Channel
Material (core)	Alumina (Al_2O_3)
Material (flange)	Hastelloy/ PEEK / PPS
Design temperature (° C)	180
Design pressure (MPa)	2
Internal volume (ml)	max. 144

not the apparatus can be used in the actual operation environment by clarifying the permissible temperature difference between the temperature of the fluid supplied to the temperature adjustment channel at the startup and the surface temperature of the apparatus. The permissible temperature difference between the apparatus and fluid that would ensure the safe use of the apparatus was set to 65℃ on the basis of the results of thermal shock tests and analysis using steam and liquid nitrogen. If the temperature difference between the apparatus and fluid is expected to exceed 65℃ at the starting up of the apparatus, the apparatus must be preheated step by step to manage the temperature difference with the fluid.

In the future, actual operational data will be accumulated to further improve the accuracy of the permissible values and to expand the applications of the apparatus for commercial production.

4. Implementation of Consignment testing

In the early days of the SMCR™ development, it was intended to have customers optimize the reaction conditions applied to the SMCR™, and Kobe Steel aimed at commercialization by designing the SMCR™ in accordance with the reaction conditions. There was, however, a problem in that it is difficult for customers to prepare experimental equipment and determine experimental conditions. Now, as Kobe Steel's latest effort, the company is happy to accept the consignment of various kinds of tests in the stages of basic testing and bench-scale testing. This owes to the fact that Kobe Steel has basic testing equipment and a bench unit in its laboratory. In addition to the basic testing and bench-scale testing, the specifications of the SMCR™ for commercial production are examined on the basis of the test results, and the economic evaluation is carried out in a consistent manner, which raises the expectations for promoting commercialization.

4.1 Bench unit

A bench unit was installed in 2018 at Kobe Steel's Corporate Research Laboratories. The appearance of the bench unit is shown in **Fig. 7**. The bench unit has the devices and instrumentation (tank, pump, various sensors, separator, heat medium/refrigerant supply unit, etc.) necessary for the SMCR™ operation and is configured to adapt the SMCR™ in accordance with reaction conditions and throughput. The major purposes of installing/using the bench unit are as follows:

① To acquire knowledge about operation and control on a commercial scale by conducting bench-scale testing incorporating the SMCR™.
② To obtain complementary data between basic testing and commercial production through bench-scale testing using the SMCR™.
③ To provide a showroom of unit equipment for flow synthesis, and to carry out external promotion.

4.2 Verification test using bench unit

Verification testing was performed using the

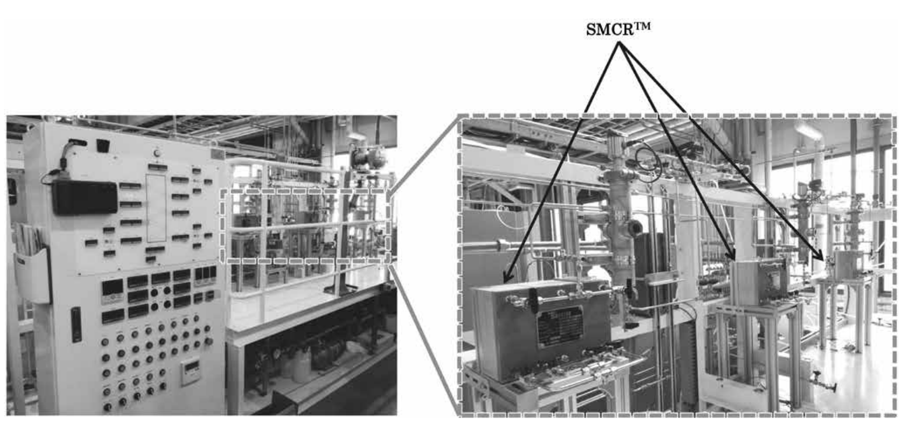

Fig. 7 Bench stage unit

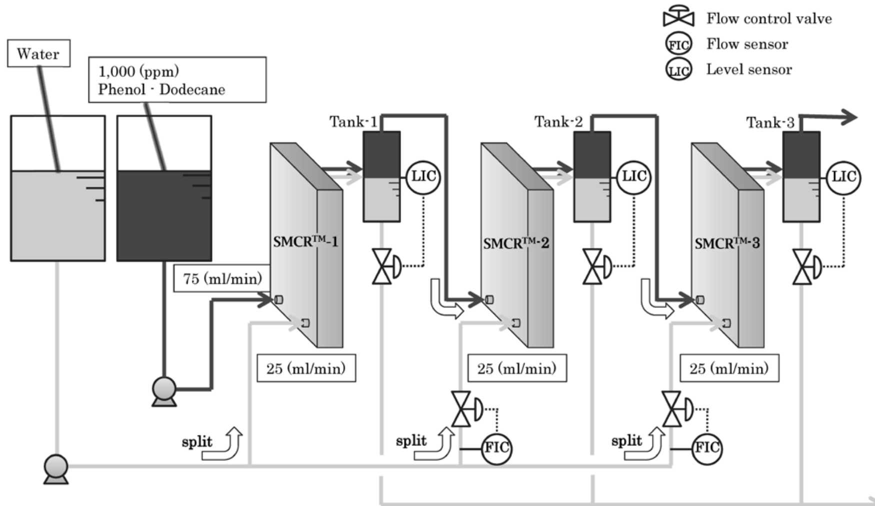

Fig. 8 Flow diagram of SMCR™ bench unit

bench unit to confirm the usefulness of the bench unit installed and of the SMCR™ incorporated therein. This test targeted the extraction operation, in which dodecane with phenol, dissolved as a simulated extraction substance, and water, a phenol extractant, were brought into liquid-liquid contact in an SMCR™ so that phenol was extracted to the water side. The data obtained from the extraction test in a single channel was used to estimate the performance of the standard type SMCR™ to confirm whether the estimated extraction efficiency could be achieved. **Fig. 8** shows the flowchart of this test, and **Fig. 9** shows the appearance of the standard SMCR™ installed.

The standard type of SMCR™ has 24 semicircular channels with a radius of 1 mm. This extraction operation was carried out in a multi-stage (3 stage) operation system, in which dodecane with 0.1 wt% phenol dissolved was pumped sequentially from the 1st stage to the 3rd stage, while solvent clean water was supplied to each of the stages. A level sensor was attached to the phase separation tank downstream of the SMCR™ at each stage to measure the interface position between the phenol-dodecane solution and water in the tank. The adopted method includes controlling the discharge amount of the water phase so that the interface position is kept constant while pumping dodecane to the SMCR™ in a later stage, thereby enabling simple operation without increasing the number of pumps.

The results of the bench-scale testing are shown in **Fig.10** to **Fig.13**.[4] Fig.10 shows the temporal change in the height of the dodecane-water interface in each phase separation tank attached to the subsequent stage of the respective SMCR™. These figures confirm that the height of the dodecane-water interface in the phase separation tank in

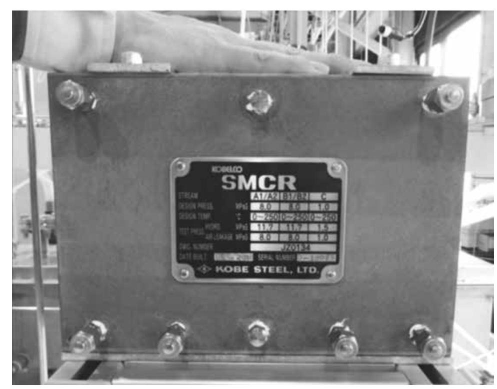

Fig. 9 Standard SMCR™

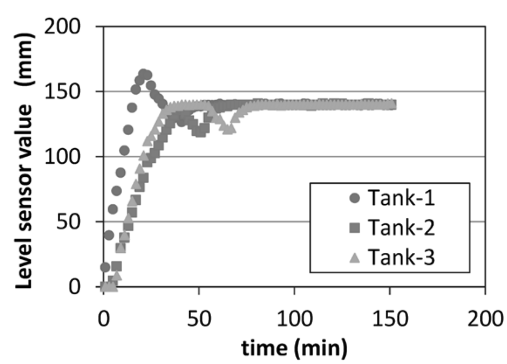

Fig.10 Temporal change of interface level at each stage

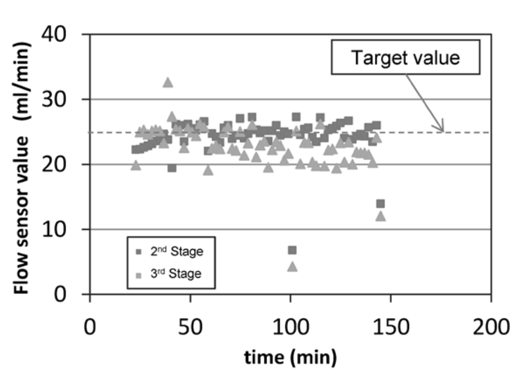

Fig.11 Flow rate of water phase of each stage

each stage was successfully kept constant. **Fig.11** shows the temporal change in the amount of water supplied to each stage, confirming that the distribution of water to the 2nd stage and the 3rd stage can be controlled stably near the set value, although there is some variation.

Fig.12 shows the extraction efficiency of phenol in dodecane to the water side at each stage. The efficiency E of phenol extraction is expressed by Equation (1) using the initial phenol concentration C_0 in dodecane and the phenol concentration C_n in dodecane at the outlet of the phase separation tank at the nth stage:

$$E=\frac{C_0-C_n}{C_0} \quad \cdots\cdots (1)$$

Fig.12 shows that the extraction efficiency of each stage is constant regardless of the passage of time, allowing stable extraction.

Fig.13 shows the experimental and estimated values of phenol extraction efficiency at each stage. The estimated value of phenol extraction efficiency in each stage is the equilibrium extraction efficiency calculated from the relationship of phenol concentration between the water side and the dodecane side (extraction isotherm) in the equilibrium state, in which the relationship has been obtained by a batch-type extraction test on a beaker scale, Fig.13 shows that the estimation results based on the equilibrium-state phenol-extraction data obtained by the batch-type extraction test and the bench-scale testing results are almost the same, confirming that an SMCR™ extraction system can be designed by using the batch-type extraction test results.

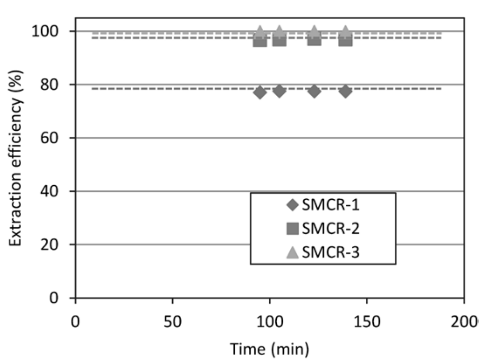

Fig.12 Temporal change of extraction efficiency

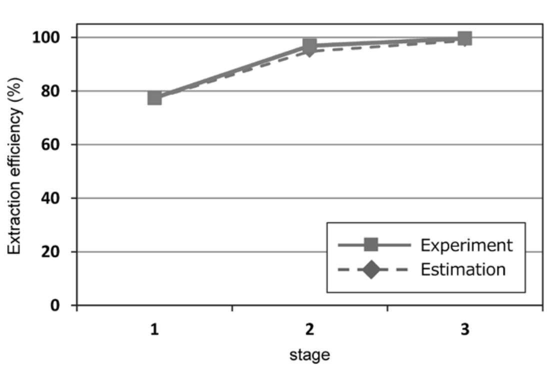

Fig.13 Experimental and estimated values of phenol extraction efficiency at each stage

5. Future perspective of SMCR™

In efforts such as the apparatus supply of SMCR™ and the latest basic/bench-scale testing, the goal is to achieve the early commercialization of SMCR™, and the standardization of SMCR™ units will be carried out with a mid- to long-term vision. Currently, custom-made specifications of SMCR™ for commercial production are being considered on the basis of the results of basic testing for each application and each customer. Cost reduction will be realized by defining a standard unit including the pump, tank, and instrumentation in addition to SMCR™ and by lining them up in accordance with the throughputs.

On the other hand, in order to apply the standard unit to processes with different reaction conditions, a circulation system as shown in **Fig.14** will be built as a standard unit. This is the concept of, for example, in the cases of multi-stage extraction, returning the ingredients and solvent to their respective tanks after extraction for a certain period of time in an SMCR™, and repeating this operation for a certain period of time to replenish the solvent to continue the extraction operation in the SMCR™. This operational method eliminates the need to change the specifications of the unit for any extraction time and number of extraction cycles, thus simplifying the configuration of the unit. In the future, detailed specifications and verification tests will be conducted on these standard units with the aim of putting them into practical use for their early commercialization.

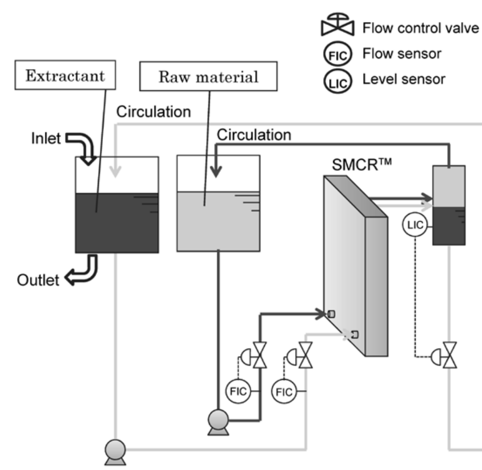

Fig.14 Concept of circulation unit

Conclusions

This paper has introduced the characteristics of the SMCR™, as well as Kobe Steel's latest efforts and future perspectives. The recent measures taken against environmental problems and the growing need for higher production efficiency will make a transition from batch processing to flow synthesis production. Such process conversion, however, has many problems that must be solved. Hence, Kobe Steel has been conducting the development of apparatuses such as a ceramic SMCR™, along with basic and bench-scale testing, so as to promote customer adoption of flow synthesis. As a result, there are an increasing number of cases where specific examination is conducted with the awareness of commercialization. To this end, Kobe Steel hopes to contribute to the conversion to flow synthesis production by achieving early commercial production and accelerating the initial examination of flow synthesis through the supplying of standard units.

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