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CFD-based Design of Multi-tube Heat Exchange Type Compact Reactor

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The production capacity of compact reactors with micrometer or millimeter-scale channels or tubes is increased by numbering-up. In previous studies, a multi-channel plate type reactor and a multi-tube type reactor (MTR) were developed and applied to extraction and reaction operations. Fluid distribution has often been evaluated to design these reactors, but temperature control, which is critical to the reaction, has not been fully considered. It is important to solve this problem and establish a design method. In this study, computational fluid dynamics (CFD)-based design was performed so as to achieve the uniform flow and temperature distributions among the reaction tubes in the MTR, where an exothermic reaction proceeds in each tube with an immobilized catalyst and the reaction temperature is controlled by a coolant flowing outside the tubes. Effects of multi-tube arrangement of lattice, concentric circles and single circle, shell cross-sectional shape of circle, rectangle and ring, and reaction tubes with or without catalyst-free inert sections on the reactor performance were investigated by CFD. The usefulness of a two-step approach of designing the MTR after designing the double-tubular reactor was confirmed through a case study on parallel reactions.

1. Introduction

Compact reactors with micrometer or millimeter-scale channels have been attracting attention in the fields of fine chemicals and pharmaceuticals in recent years due to their advantages of efficient mass and heat transfer and precise control of short residence time (Tsubogo et al., 2015). The production capacity of compact reactors is increased by numbering-up, which means the parallelization of channels or tubes. Typical compact reactors with numbering-up structure are multi-tube type reactors (MTRs) and multi-channel plate type reactors (MCPRs). The MTR can be regarded as a scaled-down version of the conventional shell-and-tube type device. The MCPR has a structure in which parallelized channels are processed in one plate and the plates are stacked as necessary.

Flow distributions in the compact reactors with numbering-up structure for single-phase and multi-phase flows have been evaluated by experiments and simulations (e.g., Rebrov, et al., 2007; Shen, et al., 2018; Park et al., 2018), and the fluid distribution section in the reactors has been designed to achieve the uniform flow distribution (e.g., Dong, et al., 2017; Tonomura, et al., 2019a; Tonomura, et al., 2022). Applications of the compact reactors to reaction and extraction operations have also been reported (e.g., Noishiki et al., 2013; Su et al., 2016; Tonomura, et al., 2019b). However, temperature control, which is critical to the reaction, has not been fully considered, especially for the MTR. It is important to solve this problem and establish a design method. In this study, computational fluid dynamics (CFD)-based design is performed so as to achieve the uniform flow and temperature distributions among the reaction tubes in the MTR, where an exothermic reaction proceeds in each tube with an immobilized catalyst and the reaction temperature is controlled by a coolant flowing outside the tubes.

2. CFD-based two-step design for MTR

Figure 1 shows an example of MTRs. The MTR consists of a reaction fluid distribution section, a multi-tube section, and a reaction fluid collection section. The multi-tube section consists of parallelized reaction tubes and a shell surrounding them. The structure and dimensions of the MTR have to be determined so as to satisfy given

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design conditions and requirements such as a reaction system, reaction rate, target production volume. In this study, a two-step design using CFD is proposed to determine them. The proposed two-step design is as follows: Step 1 is the design of a double-tubular reactor with reaction and coolant fluids flowing through the inner and outer tubes, respectively, as shown in Figure 2. Here, the design and operation variables are determined using CFD model. Step 2 is the design of the MTR. The required number of tubes, which are parallelized in the MTR, is obtained by dividing the target production volume by the production volume of the double-tubular reactor designed in step 1. Design and operation variables such as the tube arrangement, the shape and size of the shell, the diameter and position of the coolant nozzles, and the coolant flow rate are determined using CFD model such that the temperature distribution of each tube in the MTR is equal to that obtained in Step 1. After designing the multi-tube section, the reaction fluid distribution and collection sections are designed to achieve the uniform flow distribution of the reactant.

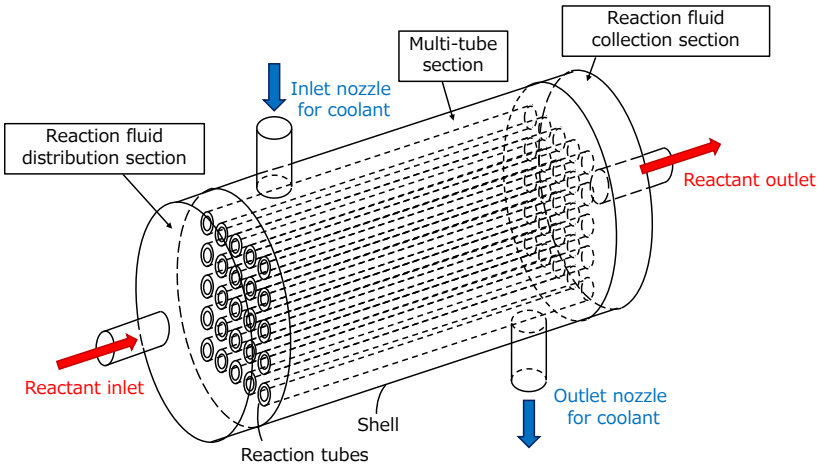

Figure 1: MTR (tube arrangement: lattice, shell cross-sectional shape: circle)

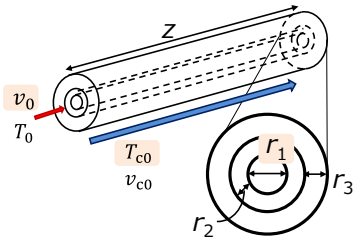

Figure 2: Double-tubular reactor

3. Case study

The usefulness of the CFD-based two-step design is verified through a case study.

3.1 Design conditions and requirements

The parallel exothermic reaction is assumed to proceed in a reaction tube with an immobilized catalyst. A is the raw material, B is the main product, and C is the by-product. These substances are dissolved in solvent D. The temperature dependence of each reaction rate follows the Arrhenius equation. The reaction parameters including standard reaction enthalpy ($\Delta_r H_i^\circ$) are shown in Table 1. The higher the reaction temperature, the more by-product is produced. The physical properties of the reaction fluid and the reaction tube are shown in Table 2. They are all assumed to be constant. This assumption does not reduce the accuracy of design results for double-tubular reactor and MTR with the liquid-phase catalyzed reaction, because the effect of temperature dependence of physical properties on the reaction rate, reaction time, and flow distribution is small that it can be ignored. In this case study, the target production volume is set to be 1.35 kg/day, and the purpose is to design an MTR that satisfies it.

Reaction 1	$\mathrm{A} \rightarrow \mathrm{B}$	$r_1 = k_{10}\exp\{-E_1/(RT)\}\ C_\mathrm{A}$
Reaction 2	$\mathrm{A} \rightarrow \mathrm{C}$	$r_2 = k_{20}\exp\{-E_2/(RT)\}\ C_\mathrm{A}$

Table 1: Reaction parameters

k_{10}[1/s]	2.5×10^{8}
k_{20} [1/s]	2×10^{18}
E_1 [kJ/mol]	60
E_2 [kJ/mol]	120
$\Delta_r H_1^\circ$ [kJ/mol]	80
$\Delta_r H_2^\circ$ [kJ/mol]	50

Table 2: Physical properties of reaction fluid and reaction tube

Density [kg/m^3]	700 (fluid), 3940 (tube)
Specific heat [J/kg/ K]	2000 (fluid), 0.5 (tube)
Thermal conductivity [W/m /K]	0.1 (fluid), 17 (tube)
Viscosity [kg/m/s]	0.0003 (fluid)
Diffusivity [m^2/s]	1×10^{-6} (fluid)
Molar mass [g/mol]	50 (fluid)

Table 3: Fixed design and operation variables for the double-tubular reactor

r_2 [mm]	r_3 [mm]	z [mm]	T_0 [K]	v_{c0} [m/s]	Mole fraction of raw material A
0.2	0.3	500	323	0.1	0.5

Table 4: Optimization result

r_1 [mm]	v_0 [mm]	T_{c0} [K]
0.72	0.01	303

3.2 Step 1: double-tubular reactor design

The design problem of a double-tubular reactor shown in Figure 2 is formulated as follows: The objective is to maximize the profit calculated by subtracting the cost of separating component C from the sales of component B. The selected optimization variables are the tube inner diameter (r_1), the inlet velocity of the reactant (v_0), and the inlet temperature of the coolant (T_{c0}). Other variables are assumed to be fixed values as shown in Table 3, and the outer wall of the reactor is assumed to be adiabatic. The upper and lower limits of the optimization variables are as follows: $0.4 \le r_1 \le 1.6$, $0.01 \le v_0 \le 0.5$, T_{c0} = 283, 293, 303, 313, and 323. The result of CFD-based optimization is shown in Table 4. At this time, the molar fractions of components B and C at the reactor outlet were 0.220 and 0.106, respectively.

3.3 Step 2: MTR design

Since the production volume of the double-tubular reactor designed in the previous section was 54.3 g/day, the number of reaction tubes required to achieve the target production volume is 25. In the MTR design, the objective is to achieve the same reaction field as the double-tubular reactor, and the design result is evaluated on the basis of the temperature distributions as well as the root-mean-square error (RMSE) between the temperatures in the flow direction of the double-tubular reactor and the MTR, which are obtained by CFD simulation (ANSYS Fluent®). First, the multi-tube section is designed assuming uniform flow distribution of a reactant to the reaction tubes, and then the reaction fluid distribution and collection sections are designed. With reference to conventional shell-and-tube heat exchangers, two candidates for the multi-tube arrangement were set: lattice and concentric circles, and two candidates for the shell cross-sectional shape were set: rectangle and circle. After combining these candidates (e.g., [lattice]x[circle]), the remaining design variables, such as the distance between the tubes and the inner diameter and length of the shell, have to be determined. For simplicity, the positions of the inlet and outlet nozzles were fixed at 0.5 mm from the end of the shell, and the inner diameter of both nozzles was fixed at 1.12 mm. In addition, it was assumed that the linear velocity of the coolant in the shell is equal to that determined in Step1 and that the total flow rate of the coolant in the shell is equal to the coolant flow rate of step 1 multiplied by the number of reaction tubes. These assumptions were used to determine the shell size.

Figure 3a and 3b show a cross-sectional view of the MTR with [lattice]x[circle] structure and the fluid temperature distributions in some representative reaction tubes, respectively. It can be seen that the temperature distributions of all tubes deviate from the result of Step 1 and the magnitude of the deviation is greater near the

center of the shell, resulting in large variations in the temperature distribution. The value of the evaluation function was 4.82 K. Similar result was obtained for the MTR with [concentric circle]x[rectangle] structure. Figures 4 and 5 show the result of the MTR with [concentric circles]x[circle] structure and [lattice]x[rectangle] structure, respectively. It was shown that the fluid temperature distributions of both results were improved compared to the [lattice]x[circle] case. The values of the evaluation function for [concentric circles]x[circle] and [lattice]x[rectangle] structure were 0.52 K and 0.55 K, respectively. Although these values were small, it was seen that the temperature is already non-uniform around the inlet of the reaction tubes and that there are tubes with different temperature variations around the outlet of the reaction tubes. Considering from the CFD simulation results that these were caused by the coolant flow patterns at the tube inlet and outlet, catalyst-free inert sections with lengths of 200 mm and 50 mm were installed before and after all tubes in the MTR with [concentric circles]x[circle] structure, which showed the best result so far, respectively. As a result, although different temperature variations around the tube outlet were no longer observed, the uniformity of the temperature distribution among the parallelized tubes was not improved, and even if the radii R_1 and R_2 were adjusted, there was no effect. Finally, when the [single circle]x[ring] structure shown in Figure 6a was investigated as a multi-tube arrangement different from the conventional ones, a uniform temperature distribution was obtained as shown in Figure 6b. At this time, the value of the evaluation function was 0.11 K.
Following the design of the multi-tube section with the [single circle]x[ring] structure, the reaction fluid distribution and collection sections were designed to achieve the uniform flow distribution of the reaction fluid. A symmetrical shape was assumed for the fluid distribution and collection sections in this study. As shown in Figure 7, a manifold-type fluid distribution device (Park et al., 2018) was designed. The shape of the fluid distribution device was a cylinder with a diameter equal to the shell diameter of the previously designed multi-tube section, and the reactant was supplied from the inlet nozzle at a velocity of 0.086 m/s. The length of the cylinder-type fluid distribution device was set to 5 mm by trial and error. As a result of CFD, it was confirmed that the reactant was evenly distributed to the reaction tubes and the flow velocity of each distributed reactant was 0.01m/s, which is equal to the design result of the multi-tube section.

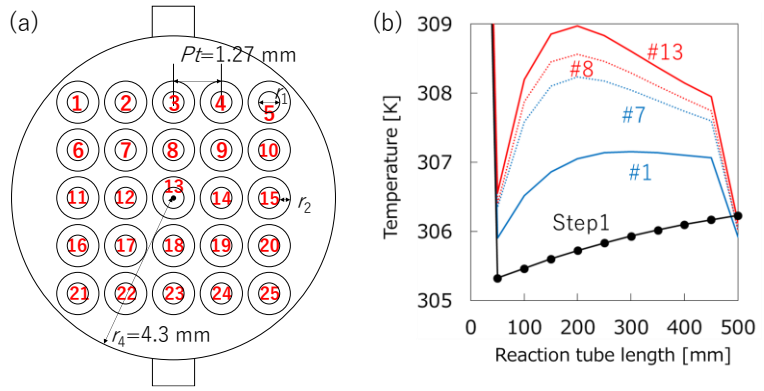

Figure 3: MTR with [lattice]x[circle] structure

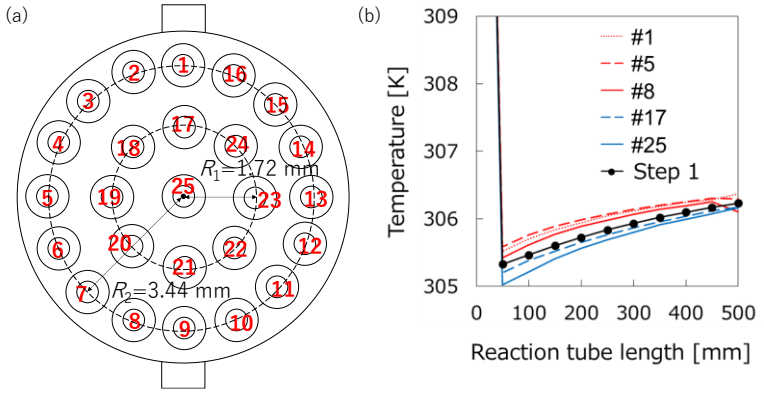

Figure 4: MTR with [concentric circles]x[circle] structure

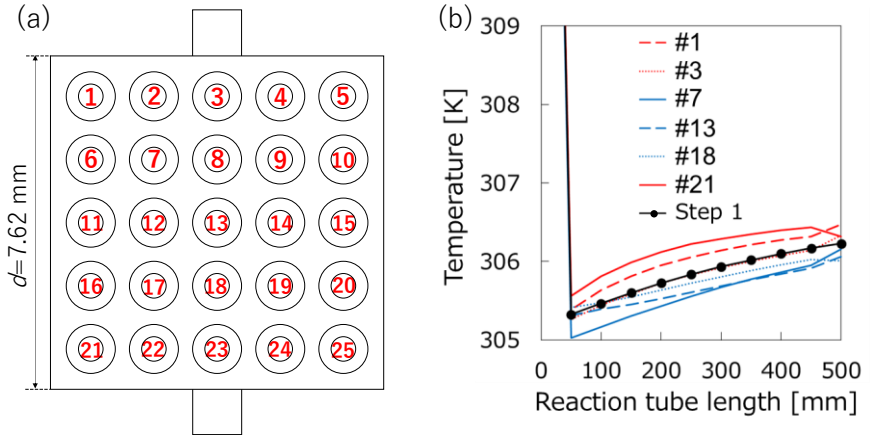

Figure 5: MTR with [lattice]x[rectangle] structure

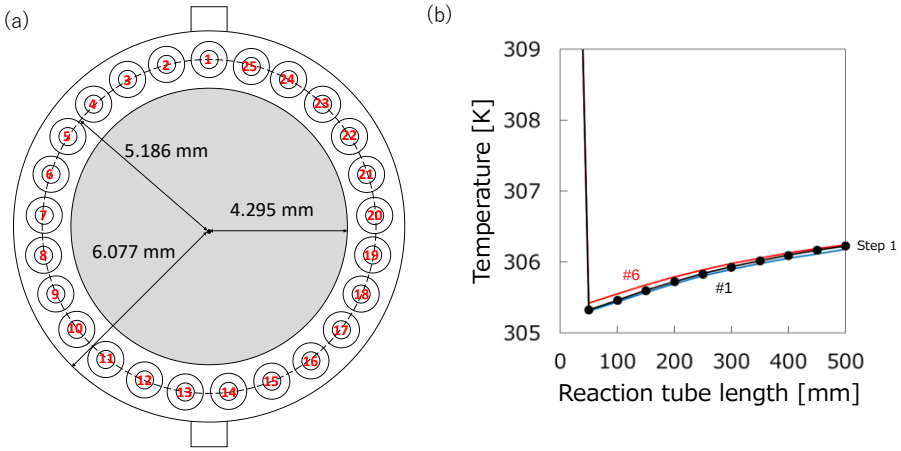

Figure 6: MTR with [single circle]x[ring] structure

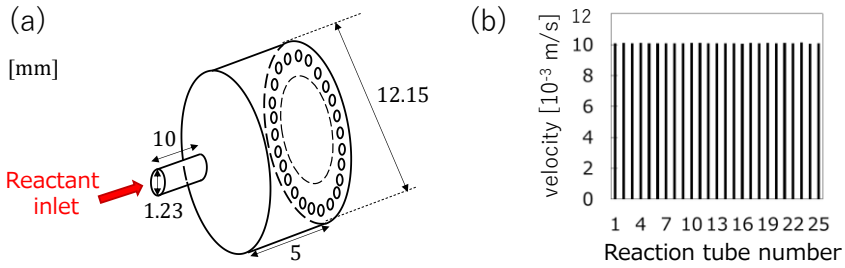

Figure 7: Fluid distribution section design result

Finally, when calculating the mean square error between the mole fractions of component C at the outlet of the reaction tube in the MTR and the reaction tube in Step 1, [lattice]x[circle] structure is 6.01 x 10^{-3}, [concentric circles]x[circle] is 6.27×10^{-5}, and [single circle]x[ring] structure is 7.77 x 10^{-6}. It was demonstrated that a stepwise design based on CFD can derive an MTR that achieves an increase in production while reducing the deviation from the reaction results of a single tube.

4. Conclusions

In this study, CFD-based design is performed so as to achieve the uniform flow and temperature distributions among the reaction tubes in the MTR, where an exothermic reaction proceeds in each tube with an immobilized catalyst and the reaction temperature is controlled by a coolant flowing outside the tubes. The main results are as follows. CFD results of the MTR under concurrent heat exchange operation suggested that the flow pattern of coolant in the shell is an important factor for precise control of temperature distributions in the tubes. It was shown that the installation of inert sections, which are not filled with catalyst, before and after each reaction tube leads to the uniformity of the heat exchange operation with the coolant in all tubes. Furthermore, in order to minimize the temperature deviation among the reaction tubes, a device structure was proposed in which the shell shape was ring and the reaction tubes were arranged in a circle. As a result, the temperature deviation of the proposed device structure was 1/5 of that of the conventional device structure.

Nomenclature

C – concentration, mol/m^3
E – activation energy, kJ/mol
k – reaction rate constant, 1/s
R – gas constant, J/mol/K
r_1 – tube inner diameter, mm
r_2 – tube thickness, mm
r_3 – double tube gap, mm
T_0 – inlet temperature of the reactant, K
T_{c0} – inlet temperature of the coolant, K
v_0 – inlet linear velocity of the reactant, m/s
v_{c0} – inlet linear velocity of the coolant, m/s
z – tube length, mm
$\Delta_r H_i^\circ$ – standard reaction enthalpy, kJ/mol

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