Multifunctional exchanger-reactor: a design, fabrication and experimental study

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Résumé - Multi-functional equipments are particularly interesting for various processes with the aim of process intensification. Miniaturization of process equipment enhances transfer phenomenon, and is one of the future development trends for chemical process and energy industries. This paper introduces a miniaturized design of multi-functional heat exchanger-reactor. Two exchangers are fabricated using rapid prototyping. Heat transfer experiment shows good heat exchange efficiency between 2500 $W \cdot m^{-2} \cdot K^{-1}$ and 5000 $W \cdot m^{-2} \cdot K^{-1}$ depending on flowrates. Mass transfer study reveals efficient micromixing effect, being comparable with some micro-structured reactors in the literature. Some practical and technical aspects are discussed regarding flexibility, material constraints, etc.

Nomenclature

A_o	heat transfer surface area, m^2	U	heat exchange coefficient, $W \cdot m^{-2} \cdot K^{-1}$
C	concentration, mol/L	w	channel width, m
F	correction factor, --	V	internal volume, mL
h	channel height, m	*Symboles grecs*	
l	length, m	Φ	exchanged heat power, W
ΔP	pressure loss, Pa	*Indices et exposants*	
Q	volume flow rate, mL/min	h, c	hot/cold side
Re	Reynolds number, --	i, o	inlet or outlet of fluid
T	temperature, °C	k	chemical species
t_m	micromixing time, s	t	tube side

1. Introduction

High efficiency equipments are usually desired by processes like chemical reaction, thermal energy generation and conversion, etc. For a chemical reaction, a well adapted reactor may give high yield, simplifying post purification processes and decreasing wastes. In the case of a heat exchanger, low thermal resistance between the hot and cold side may reduce energy consumption. Generally, most designs that give efficient transfer and render high conversions may be included in the domain of process intensification [1].

One of the techniques to realise process intensification is through miniaturization - more precisely through using equipments with locally miniaturized structures. Heat and mass transfers may be enhanced since the *surface-to-volume* ratio is raised by several times. Some millimetric devices have been designed for industrial process applications while micrometric designs have been realized for analytic instrumentation and the so-called *lab on a chip* [2].

The concept of multifunctional device is widely accepted by both academic and industry. Highly exothermic chemical reactions should be processed with an appropriate thermal management system. This is aimed at not only gaining a high conversion of reaction, but also at avoiding hot spot formation, clogging or even runaways. Micro-structured reactors (MSR) which integrate thermal control (heat management) function in the reactor could help to reach the goal. While for extremely exothermic reactions, especially those very rapid ones that

require precise thermal control, some methods like using multi-injection reactor were proposed in the literature [3, 4].

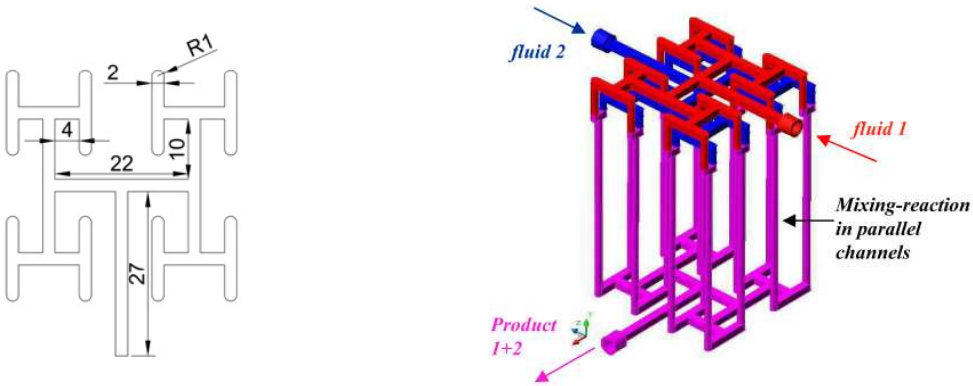
This paper introduces systematically the design of a mini-scale exchanger-reactor, its concept and fabrication. Some technologies like micro-tomography analysis are introduced as a possible fault detection technique in this sector of research.

2. Design of multi-functional exchanger-reactors

The internal numbering-up of multiple channels have been studied by several prototypes in our previous publications [5-10]. The main ideas are: i) to use arborescent structures for uniform fluid distribution; and, ii) integration of heat exchangers and reactors for a multi-functional device purpose.

Firstly, the general philosophy of arborescent component and its applications are introduced by Luo et al. 2007 [5]. Different multi-scale devices are designed such as fluid distributor, mixer and heat exchangers. Then prototypes of several typical applications have been done. In the paper of Luo et al. 2008 [6], fluid distribution characters and pressure drops are introduced and tested. The first results are that fluid distribution tends to be uniform by using arborescent structure and that pressure loss has been verified to be in a low level. Then in the specific application of cross flow heat exchanger, different configurations are tested [7]. Exergy analysis is employed for heat exchanger evaluation by Fan et al., 2009 [8]. Utilisation as mixer is studied by Fan et al. 2010 [9], showing that impinge of fluid through arborescent structure is favourable to the mixing efficiency.

Another multifunctional exchanger-reactor is introduced by Guo et al., 2012 [10], in which two fluids are mixed before their reaction with heat exchange. The reactor has two inlets of different fluids and one outlet for product. Two fluids of each inlet are injected and then distributed into 16 channels. The two fluids then contact through T-junctions where mixing happens with counter-current impingement. After, the mixture goes along the channels to gain complete mixing. The final mixture will then be collected to the outlet. Symmetry design of this nature-inspired distributing/collecting structure makes the flow path of fluid in every channel being identical. Non-uniformity of flow distribution between channels is minimized. Heat exchange chamber is located outside the vertical parallel channels. Inlet and outlet are arranged in such a way that utility fluid flows reversely with inside fluid. The schematic view of the studied exchanger-reactor is shown in Figure 1.



a: Arborescent distributor (mm) *b: An example of exchanger-reactor design*

Figure 1: *Multichannel exchanger reactor design using arborescent structure*

Two reactors with different sizes are studied. One (named as M2) has a channel diameter of 2 mm while the other reactor (M1) is more compact with its channel diameter being 1 mm. For better visualization of the flow field, the cross-section of all channels of distributors and

collector is square shape (2×2 mm for M2 and 1×1 mm for M1), as indicated in Figure 1a. Total internal volume of reactor M1 and M2 is 1.27 mL and 6.07 mL, respectively.

3. Fabrication

Several rapid prototyping technologies have been served in the production of mini processing equipments. In this section we firstly introduce two main technologies we have employed, i.e. DMLS and SLA. Then we present the prototypes of the exchanger-reactor.

3.1. Brief introduction to rapid fabrication technologies

3.1.1. Stereolithography

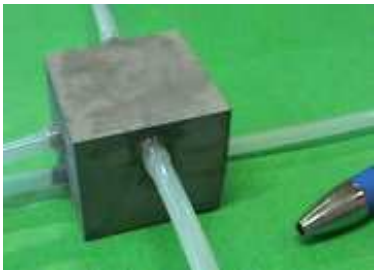
In SLA, photopolymer is cured selectively at the local focus of ultraviolet laser. The process is carried out layer by layer and a supporting platform ascends once a certain layer is completed. Precision of product depends on the layer depth and in our case the layer depth is 100 µm. The smallest diameter of channel should be no less than 1 mm, considering the fragility and connections [11]. With post processing, the quality of final product (surface planarity) is guaranteed.

Polymer used in the construction of compact heat exchanger is advantageous in less weight, high resistant to chemical corrosions, and low cost. Low thermal conductivity may result in low global heat transfer coefficient compared with metal made heat exchangers. High temperature processes should be avoided too since polymer generally does not support a temperature higher than 100 °C. A review of polymer compact heat exchangers is found in the paper of Zaheed and Jachuck (2004) [12].

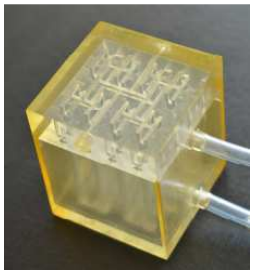
3.1.2. Direct Metal Laser Sintering

DMLS is used to the fabrication of metal compact exchanger-reactor. DMLS is to frit small sized metal powders in a selective manner by using laser beams. Powders generally have a particle diameter of 10-45 µm, and the layer depth is 50 µm [13], which makes the product more precise in resolution compared with that of SLA. Several materials like alloy steel, aluminium and cobalt-chrome could be used. Depending on the materials, the part could resist a temperature as high as 1200 °C [13, 14]. So the final part is more functional (production) than demonstrative (prototyping).

Both DMLS and SLA method require consideration on supporting issue of internal structures. Powders or polymer liquid should be able to be evacuated after the fabrication process.



a: M1, DMLS, Cobalt-Chrome



b: M2, SLA, Clear resin

Figure 2: *Exchangers fabricated by DMLS (M1) and SLA (M2)*

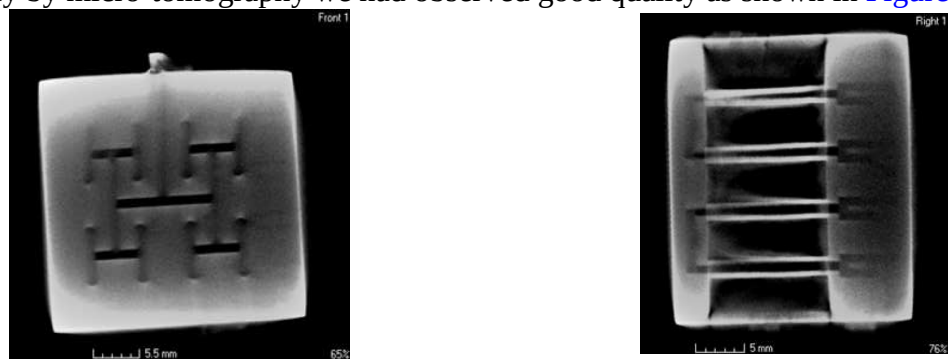
3.2. Fabrication of exchangers with DMLS and SLA

Both DMLS and SLA are used for the fabrication of designed exchanger-reactors. In the case of M1, Cobalt Chrome is used as the material used by DMLS. While in the case of M2,

transparent resin is used for SLA so that several experiments with optical observation are possible. The photos of reactors are shown in Figure 2.

3.3. Quality issue

Rapid prototyping like SLA and DMLS usually requires quality inspection after fabrication. Transparent parts fabricated using SLA could be simply inspected by eyes; while for metal parts made by DMLS, internal structure details could be inspected with micro-tomography analysis. Most frequently, with the SLA fabrication, we encounter faults caused by thermal expansion. For long channels with a fine tube thickness, expansion in longitude direction usually results in bending. To avoid expansion deformation, supporting additions are recommended. In the case of DMLS, fabrication fault usually happens in the post-sintering process. Since material becomes homogenous during this procedure, when some minor deformation may happen. Usually this deformation is within an acceptable tolerance and fortunately by micro-tomography we had observed good quality as shown in Figure 3.



a: Top view - distributor *b: Lateral view - channels*

Figure 3: *Tomography for fault detection of a metal mini-exchanger fabricated by DMLS*

4. Experimental evaluation on heat exchange and mixing

4.1. Heat exchange study

4.1.1. Experiment system

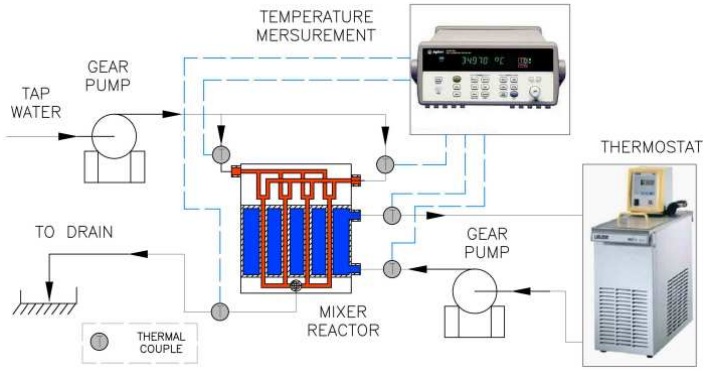


Figure 4: *Heat exchange experiment schematic*

The experimental setup is composed of two loops: process fluid loop and utility fluid loop. Process fluid is pumped through distributor, channels (tube side) and collectors; while utility fluid is taken as heating loop, and it is located at the shell side of heat exchanger. Two adjustable pumps (gear type, max. 2000 $mL \cdot min^{-1}$, Diener Precision Pump) were used to circulate the two loops of fluid. A circulation thermostat (LAUDA E200) was used to control the temperature of utility fluid. Thermal couples (K-type) were used for temperature measurements. A data acquisition centre (Agilent 34972A) recorded the temperatures

automatically. Process loop through channels was circulated with tap water, whose temperature was quite stable during the test. The heat-transfer (utility) fluid flowed through the thermostat by obtaining a stable high temperature. Massflow of each fluid was related with the rotational speed of gear pump.

Schema of the experimental setup is shown in Figure 4. Globally the heat exchanger and its connecting tubes were insulated from the ambient during the experiments. Thermal couples were placed inside the connecting tubes as close to inlet/outlet port as possible. All experiment data were captured when temperatures were stable. A repeatability study was done with consistent results been found.

4.1.2. Calculation of heat exchange coefficient

Global heat transfer coefficient is calculated with measured heat exchange power, heat transfer surface area (channel outside area) and mean temperature difference:

$$U_{experiment} = \Phi / A_o F \Delta T_m \quad (1)$$

In the equation, A_o stands for the outside surface area of channels, m^2; F is the correction factor for using the logarithmic mean temperature difference (LMTD) method. Here we consider our heat exchanger-reactor as counter-current configuration with F=1. The log-mean temperature of LMTD is calculated by equation:

$$\Delta T_m = \frac{\Delta T_2 - \Delta T_1}{\ln(\Delta T_2 / \Delta T_1)}, where: \Delta T_1 = T_{h,i} - T_{c,o}; \Delta T_2 = T_{h,o} - T_{c,i} \quad (2)$$

ΔT_1 and ΔT_2 are temperature differences at hotter and colder end of exchanger, respectively.

4.1.3. Results

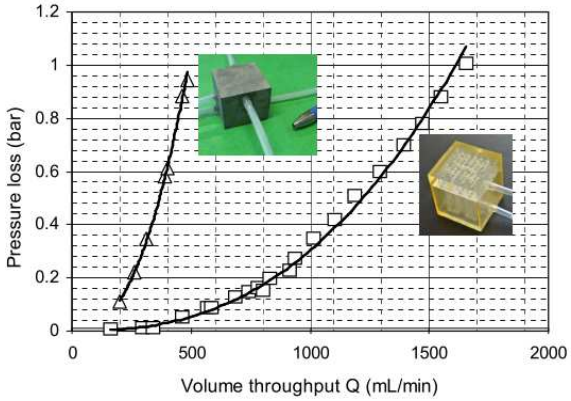


Figure 5: *Pressure loss characters for M1 and M2*

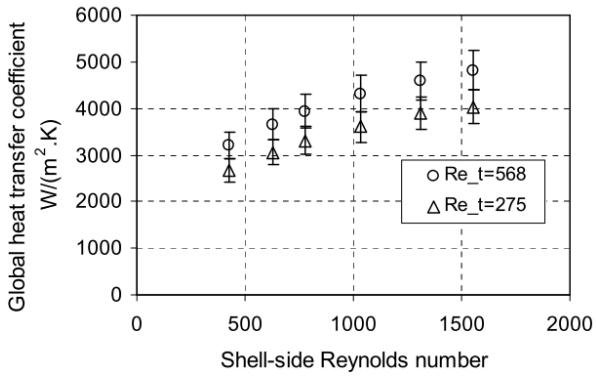


Figure 6: *High heat exchange coefficient obtained from experiment, M1*

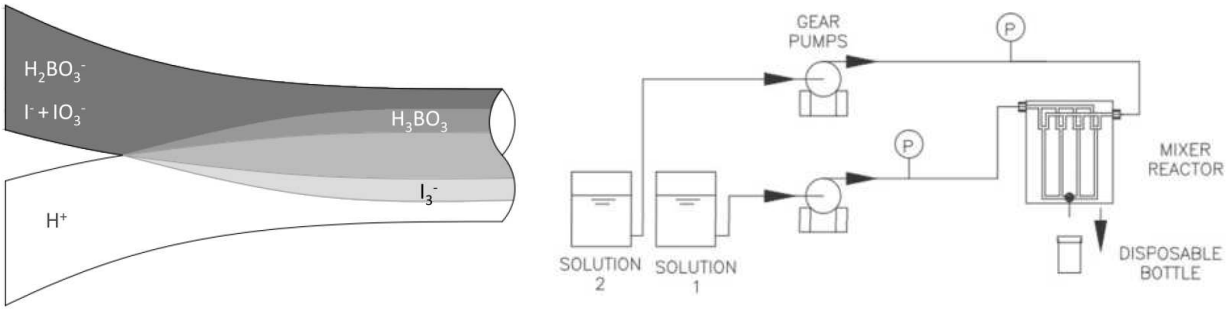
Pressure loss characteristics are tested firstly, which is shown in Figure 5. Throughputs of M1 could reach 500 mL/min while that of M2 is 1600 mL/min, for a pressure loss under 1 bar. Global heat exchange coefficients (U) under tested conditions, shown in Figure 6, are at a quite high level. Values of experimental global heat exchange coefficients varies around 2500~5000 $W \cdot m^{-2} \cdot K^{-1}$. High flowrates of utility/process fluid result in high heat transfer coefficient.

4.2. Micromixing study

4.2.1. Experiment method and setup

Competitive iodide/iodate reaction is used in this study to evaluate the micromixing performance. This reaction scheme is sensitive to mixing in molecular level by the production

of iodine (I_2). Two proton-competing parallel reactions include a neutralization reaction (R1) and a redox reaction (R2). Basic principle of iodide/iodate reaction is shown in Figure 7a. The selectivity of parallel reactions depends strongly on mixing characteristic time and reaction time. Globally excessive $H_2BO_3^-$ could consume H^+ rapidly since the reaction (R1) is quasi-instantaneous. However at molecular level, locally excessive H^+ may contact with iodide and iodate ions. This will produce iodine in an irreversible manner. Redox reaction is comparatively slower than the neutralization reaction. However both reactions are much faster than the mixing for current study.



a: Principle of iodide/iodate reactions *b: Experimental schema*

Figure 7: *Micromixing test principle and setup, using iodide-iodate reactions*

With the existence of iodide ions, a part of iodine could yield tri-iodide ions through equilibrium reaction (R3). Iodide, iodine and tri-iodide all exist in the final solution complex. The tri-iodide ion shows absorption peaks to ultraviolet (UV) light at wavelength of 286 nm and 353 nm, respectively. Its concentration then could be determined quantitatively by employing a spectrophotometer (HACH LANGE DR3900, software ver. 1.3).

4.2.2. Micromixing evaluation with IEM model

IEM model, developed for plug flow equipments, is used to estimate micromixing time. Applying this model requires that two solutions flowing inside a reactor have the same flowing age starting from their initial contact, which is exactly our case. Molecular exchange between two solutions is assumed to happen at a same time constant as that of mixing time t_m. Solutions are treated separately by keeping the concentration evolution of each one using differential equations in Eq. (3).

$$\frac{dC_{k,1}}{dt} = \frac{\langle C_k \rangle - C_{k,1}}{t_m} + R_{k,1}; \quad \frac{dC_{k,2}}{dt} = \frac{\langle C_k \rangle - C_{k,2}}{t_m} + R_{k,2}; \quad \langle C_k \rangle = \alpha C_{k,1} + (1-\alpha) C_{k,2} \qquad (3)$$

where R denotes the change of concentration for species k in concerned stream by reaction, mol/(L·s); t_m the exchange time constant, (same value of mixing time), s; α the global volume flow proportion of solution 1, which is 0.5 in our case. C stands for the concentration of species, mol/L.

4.2.3. Micromixing discussion

With estimated micromixing time and dissipation rate (calculated from pressure loss), we are able to compare the mixing performance of our prototypes with other micro-structured designs reviewed in [15], as shown in Figure 8. It is worth noting that micromixing times of other structures are obtained with the same iodide/iodate scheme and IEM model.

Both reactor M1 and M2 are found to be comparable in performance, under some flowrate conditions, with certain micro-structured reactors. For M2, at low dissipation rates (defined as the pumping energy consumption by a mass unit of water) around 10 W/kg, its micromixing time corresponds with that of single T-mixer with channel dimension of around 800 µm and

that of IMTEK Tangential mixer with channel dimension 300×100 µm. At higher energy dissipation rates, micromixing time of M1 is quite comparable (sometimes better than) several micro-devices, including the triangular interdigital micromixer by Mikroglas (channel dimension 50×150 µm), and Starlam micromixer by IMM (channel dimension ~100 µm).

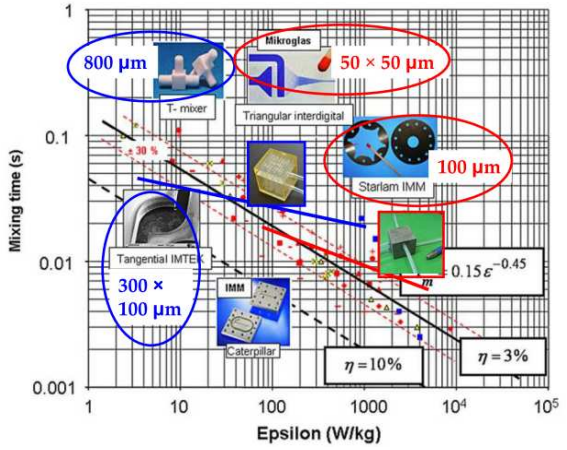


Figure 8: *Micromixing with power dissipation rate - a comparison. Figure adapted from [10]*

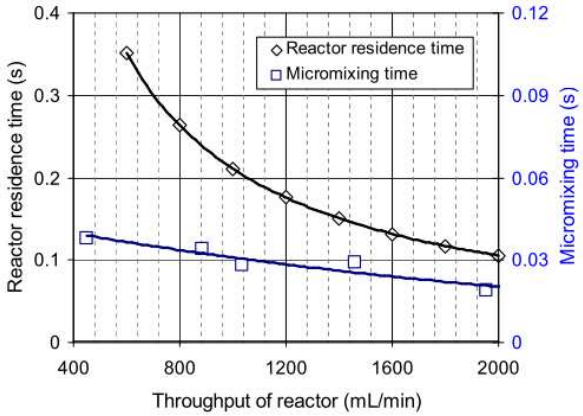


Figure 9: *Residence time changes with throughput but mixing time changes too*

5. Discussion and perspectives

Hydraulic tests show that a high throughput around 1 L/min can be realized with a pressure loss lower than 1 bar. Effective micromixing can be obtained without increasing too much pressure loss. High global heat exchange coefficients between 2500 $W \cdot m^{-2} \cdot K^{-1}$ and 5000 $W \cdot m^{-2} \cdot K^{-1}$, depending on flowrates, are achieved according to experiments. The proposed exchanger-reactor may serve as a continuous reactor for exothermic or endothermic reaction, whose residence time may be varied and whose conversion is very sensitive to micromixing.

5.1. Residence time variance - a constraint

Some inconveniences are encountered during our research, with the flexibility being one of them. Since channel lengths are fixed by initial design and fabrication, residence time through the reactor is determined. In this case varying flowrate is the only way to regulate the residence time when adapting to reactions of different speed. While at the same time, according to the mixing experiment, mixing characters is highly dependent on flowrate. Most reactions prefer a rapid mixing even if it needs different residence times due to kinetic difference. A reactor which is able to provide variable residence times but with its mixing efficiency unchanged is preferred in most processes. The studied reactors are unfortunately incapable to maintain a stable mixing efficiency, which could be seen in Figure 9.

5.2. Technical considerations

Apart from considering energy efficiency (transfer of heat and mass, flow, etc.), some constraints from technical point of view, or constraints of using modern machinery methods, should be taken into account. These reflections are recommended to be done before the designing process, and after choosing the proper fabrication technology. For example, adding supporting fins around long channels are recommended to reinforce the mechanical stability of the structure. Size of flow channels also needs to be controlled. Channels smaller than 1 mm should be fabricated using other methods instead of SLA.

5.3. Perspectives - new fast fabrication method

Electron Beam Melting (EBM) is the latest rapid fabrication technology in mini scale. Vacuum environment during the fabrication process and the utilisation of electron beam make it powerful in fabricating complicated geometries with delicate structures. Like in DMLS, metal powder is melted layer by layer. Scanning speed of EBM is two times that of DMLS. This technique, still very rare in application, is expected to provide functional final products in a 3D printing manner.

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