FLOW DISTRIBUTION AND MIXING PERFORMANCE OF A NOVEL MULTICHANNEL HEAT EXCHANGER REACTOR

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ABSTRACT
A multi-functional heat exchanger reactor (HEX) with 16 mini channels in parallel is designed, fabricated and tested. Two fluids are divided into 16 channels through distributor, then contacted at T-mixers, and finally collected. Both distributor and collector are designed following the arborescent shape. Expected advantage of using this mini HEX reactor include but not confined to: compact size, effective and fast mixing, low pressure drop, efficient and controlled heat remove/supply, easy numbering-up potential, safety, etc.

The internal fluid body is numerically analyzed using CFD approach (ANSYS Fluent) to test the flow distribution uniformity. Results show that for Reynolds number ranging from 8 to 95, good distribution uniformity (maldistribution less than 1.5 %) can be obtained. To test the mixing performance of the mini HEX reactor, Iodide-iodine reaction (Villermaux-Dushman) has been carried out. Experimental results show a good mixing at molecule scale as the flow rate increases and impingement enhanced. The design of multichannel reactor with arborescent distributing and collecting networks makes possible the numbering-up of multiple channels and thus the application of mini reactor in industry.

NOMENCLATURE

\( D_{ah} \) [-] Flow rate deviation defined by Eq. (1)
\( f_{ch} \) [kg/s] Mass flow-rate in a channel
\( f_{av} \) [kg/s] Calculated average mass flow in each channel
\( Q \) [m³/s] volume flow rate
\( V \) [m³] internal volume of reactor
\( X_s \) [-] Segregation index
\( \Delta P \) [Pa] pressure loss
\( \rho \) [kg/m³] Density
\( \varepsilon \) [W/kg] Dissipation rate defined by Eq. (2)

INTRODUCTION
In most of energy systems that involve energy production, transportation, storage and all kinds of conversions, the major components (e.g., solar collector, the boiler, condenser, absorber, adsorber, evaporator) are principally multifunctional exchanger/reactors with heat and mass transfers, with or without phase change and with or without chemical reaction. Increased interfacial flux, i.e. enhanced heat and/or mass transfer is a key issue to improve the efficiency of these systems. This approach often requires the design and development of compact and integrated thermal and fluidic components.

This necessity may be better understood when chemical reactions are involved in a certain process. The temperature control for exothermal or endothermic reactions is really important. However, many traditional designs such as stirred tank reactors that incorporate heat transfer in the process, i.e. double jacket, external or internal coil, can not supply or remove the heat almost as rapidly as it is absorbed or generated by the reaction. A compact device that combines the reaction and heat transfer into a single piece of equipment, i.e. using for instance a heat exchanger as a chemical reactor, the so-called multifunctional heat exchanger, is a good solution. In this case, the volumetric global heat transfer coefficient is higher by several orders of magnitude [1].

Secondly, when we talk about compact heat exchangers or reactors, it often concerns the channel dimension at micro-scale. The advantages of micro fluidic devices are evident such as very high efficiency and improved safety. But to obtain a comparable productivity with respect to the conventional equipments, it is obliged to put together a number of micro-mini-channels in parallel instead of mono-channel devices to increase the throughput. The so-called numbering-up issue is certainly not trivial because it brings the important problem of how to evenly distribute fluid from a single inlet port to an array of parallel micro-channels, and the reverse for
collection. The proper design of distributing and collecting networks with different dimensions is necessary. Each dimension in the network represents a specific scale.

Based on the above discussion, we summarize here the scientific constraints for the design of such fluidic and thermal components:

- Compactness;
- Uniform fluid distribution/collection;
- Effective mixing, fast chemical reaction;
- High throughput;
- Low pressure drops;
- Efficient and controlled heat remove/supply.
- Easy numbering-up

**ARBORESCENT STRUCTURE**

In “multi-scale” terms, the distributor or collector problem is topologically a connection problem between a point and a surface (or volume). The “point” is here the single inlet tube or pore, and the surface is the domain that must be fed by the distributed flow. As a result, the tree-like arborescent geometry is a “natural” option. It concerns the biomimetic method, i.e. to imitate the nature. Nature exhibits its multi-scale characteristic by the presence of tree network structures because they require the least amount of useful energy, and hence are the most efficient. It offers an impressive list of examples of equipartition or optimal allocation.

Several attempts have been made over the last decade to prompt the tree structure for the design of unit devices for chemical and energy processes. For example, a gas to liquid fractal distribution by a volume processing was introduced by Kearney [2]. Ajmera et al. [3] investigated a micro cross-flow reactor with bifurcated inlet and outlet distributors. The reactants are bifurcated into 64 inlets that feed a shallow catalyst bed. Catalyst retainers keep the bed in place while shallow pressure drop channels maintain even flow distribution across the bed. Amador et al. [4] studied manifold structures (consecutive and bifurcation) used for even flow distribution in microchannels. Senn and Poulikakos [5] also introduced tree-like channel networks as a fuel cell fluid distribution concept, which optimizes the shape of polymer electrolyte fuel cells.

However, the tree-like geometry has not been largely used for the design of multifunctional heat exchangers despite its significant advantage in numbering-up. Tondeur and his coworkers [6] proposed a geometric concept for intensification and design of the arborescent structure of fluid contactors/reactors by the topology of “channel interlacing”. However to the best of our knowledge, most of the researches are at the design stage, detailed numerical or experimental results for the performance evaluation of the multifunctional heat exchangers are not available.

**DESIGN OF HEX WITH ARBORESCENT STRUCTURE**

We propose a concept of multifunctional heat exchangers that combines fluid distributor/collector, mixer, reactor, and heat exchanger in a single and compact component with each step optimized and intensified. To do that, we used an arborescent geometry.

Figure 1 shows one example of our multifunctional heat exchanger design. Using a dichotomic logic, the first reactant (fluid 1 in Figure 1) is fed from single inlet port to 16 outlet ports that evenly distributed on a surface. Each channel corresponding exactly to the same flow-path, a uniform distribution among the parallel channels could be expected. Using the same principle, the second reactant (fluid 2 in Figure 1) is also evenly supplied. The two fluids firstly arrive in a counter-current manner and contact with each other in a “pre-mixing” chamber and then enter the stack of channels (in the form of mixture) where chemical reaction happens. If the reaction is not complete, we still have another chance to further mix the streams coming from different channels in the collector part to reinforce the mixing-reaction. By circulating the coolant around the parallel channels to evacuate or provide heat, we can control the operating conditions for the reaction, i.e. an isothermal condition, which is very interesting for various processes.

Figure 1 Schematic view of the HEX design

The example shown in Figure 1 has 16 parallel channels. We can easily imagine more channels such as 32, 64 or 128 by adding dichotomic generations in the distributor and collector parts. The length of the parallel channels is another parameter for optimization under the constraints such as residence time and reaction rate. Once this configuration is established, other structural evolutions could be expected. For example, we may use serpentine channels instead of straight channels to enhance the mixing. Other intensification techniques may also be used.
in our structure. In that case, we can modify the walls of the inner and outer channels using micro or mini-scale fins to enhance the heat and mass transfer. As for the circulation of coolant, we can use shell-type chamber as a water bath for better control of the operating temperature.

Likewise, we may design multi-fluid configurations, reactive or catalytic fluids, which will also be evenly distributed and join the on-going reaction. As shown in Figure 2, we can have as many fluids as we want simply by adding supplemental tree-like inlet structures. Moreover, the entire device can be modularized, i.e. we can link several modules in series or in parallel to achieve target productivity.

With the same geometry of Figure 1, two different sizes of HEX are studied here. One has a channel diameter of 2 mm while the other with half dimension. For clarity we named them as M1 (dia. 1 mm, length of straight channel 15 mm) and M2 (dia.2 mm, length of straight channel 50 mm), respectively. Two prototypes are then fabricated using stereolithography method. The smaller one M1 was fabricated with metal Cobalt-Chrome, while the big one M2 was fabricated with transparent resin. M1 can be used for heat exchange test because of its high conductivity of metal whereas with transparent feature of M2, we are able to visualize the flow inside with fast camera and optical tracers.

FLOW DISTRIBUTION UNIFORMITY

We present here some numerical results on flow distribution uniformity of the 3D arborescent structure within laminar flow.

The overall model and simulation was realised using different modules provided by ANSYS Workbench 12.1. The internal fluid body illustrated in Figure 1 was created and prepared for meshing. ANSYS mesh tool for CFD analysis was used to obtain the meshing, using tetra patch conforming algorithm. Near the wall, inflation of 5 layers was created for better presenting the mechanism of near wall flow transition. As grid independence study, different sizes of the mesh were tested. By adjusting the overall element number from 1.3 million, 1.7 million and 2.2 million, the comparison of calculated results between each are 5.7% and 1.8%. As a result, the overall element number was controlled to be 1.7 million after considering both speed of calculation and the precision of results.

Simulations were realised using ANSYS Fluent code. Boundary conditions were setup as velocity inlet for both inlet I and inlet II. Outlet was set to be zero static pressure. All the internal faces are defined as non-slip boundaries. For the beginning of research, we used identical inlet velocities for both inlet I and II. Pure water at constant temperature of 300K was chosen as both inlet fluids.

Laminar flow model was used as Reynolds number in the vertical channel ranges from 8 to 95. For the pressure-velocity coupling, standard SIMPLE method is used. For discretization, standard method was chosen for pressure and Second order upwind for momentum.

The solution was considered to be converged when (i) the mass flow-rate at each channel and the inlet static pressure were constant from one iteration to the next (less than 0.1 % variation) and (ii) the sums of the normalized residuals for control equations were all in the order of magnitude of 10^-6.

Bifurcation of fluid within structure of distributors and collector can be qualitatively seen from Figure 3. Within an average Reynolds number value of 95 in channels (after mixing), the flow in both distributors shows good bifurcation at each scale. Although in this case the flow along the tube is not fully developed, flow pattern after each bifurcation tends to be the same.

To better represent the distribution in a quantitative way, we defined relative flow rate deviation ($D_{ch}$) as:

$$D_{ch} = \left( \frac{f_{ch} - f_{av}}{f_{av}} \right) \times 100\%$$

$f_{ch}$: Mass flow-rate in a channel; $f_{av}$: Calculated average mass flow in each channel.

According to the definition, the distribution is perfect when $D_{ch}=0$; larger $D_{ch}$ means more maldistribution occurred among channels. Results of $D_{ch}$ for different average Reynolds number in vertical parallel channels are shown in Figure 4.

From Figure 4, it is evident that in every case of Reynolds number from 8 to 95, the $D_{ch}$ ranges from -1.50% to 1.17%, indicating the homogenous flow distribution. With the increasing average Reynolds number, the maldistribution becomes larger. This is possibly due to the bigger the Reynolds number in laminar flow, the longer it needs for fully developing. Inertial force causes maldistribution when fluid faces a bifurcation structure.
Our research teams are also carrying out experimental work on various arborescent structures. As a preliminary illustration, Figure 5 visualizes the invasion of the dichotomic tree distributor by a fluid carrying an optical tracer. The pictures are a small sample of that taken by a fast camera, and show the structure from above at different instants of time. The time span involved is a fraction of 1/15 second. These pictures qualitatively illustrate the homogeneity of the invasion.

Two gear pumps (max. 2000 mL/min, Diener Precision Pump Ltd.) are used to inject reactants inside. Flowrates can be easily adjusted by varying the input voltage and thus the motor speed. Products are collected with single-use bottles. Absorption test for quantifying the concentration of result tri-iodide were done by a UV-Visible spectrophotometer (HACH LANGE DR3900, software version 1.3), within standard plastic spectra cuvettes (1 cm optical path, single use only). The schema of the experimental set-up is shown in Figure 6.
All reactants are analytical level. Lab demineralized water is used for solution diluting. Experiments are implemented within 6 hours after solution preparation to avoid influence from ambient temperature, light, etc. Each set of experiments is implemented carefully with a repetition of at least 2 times.

We use segregation index ($X_S$) to characterize micromixing quality. $X_S$ can be calculated by the ratio of selectivity of acid during parallel reaction with that in total segregation case. Value of segregation index lies within 0 and 1, where $X_S = 1$ stands for two fluids totally segregated during mixing while $X_S = 0$ means infinitely fast mixing in molecular level.

Figure 7 shows the segregation index $X_S$ of both reactors as a function of flow-rate. It can be observed that both M1 and M2 have the same tendency, i.e., $X_S$ decreases with increasing flow-rate. Reinforced fluid impingement reduces the side redox reaction R2 so that $X_S$ becomes smaller at high flow-rates. Reducing the size of reactor increases micromixing performance when we compare the $X_S$ between M1 and M2. At around 200 mL/min until 400 mL/min, $X_S$ of M2 is about one order of magnitude higher than that of M1.

Figure 8 shows the relation of $X_S$ versus the dissipation rate defined as:

$$\varepsilon = \frac{Q \Delta P}{\rho V} \quad (2)$$

where $Q$ is the volume flow rate, $V$ the internal volume of reactor, $\Delta P$ is the pressure loss measured at the inlet and outlet and $\rho$ the density of test fluid. It can be observed that for a certain dissipation rate, the M1 has significant better mixing performance than that of M2.

It should be noted that segregation index dependents highly on the initial concentrations of each solution. In order to carry out a comparison of mixing performance between different studies using different solution concentrations, the micromixing time should be estimated, which is independent of chemical concentrations. Here we use the IEM (Interaction by Exchange with the Mean) model, developed by Villermaux and his colleagues [8]. Applying this model requires that two solutions owing inside a reactor have the same residence time (uniform distribution, plug flow), which is exactly our case. Using this model micromixing time can be related with the result from iodide/iodate reaction. Recent reviews [9, 10] on micromixing performance of different experiment results and detail description of this model are presented by Falk and Commenge.

In our study, the segregation index by reaction is used in IEM model to estimate the micromixing time. With micro mixing time and dissipation rate, we are able to compare the mixing performance of our design M1 and M2 with other configurations. Figure 9 compares our performance with that of other micro structure designs reviewed by Falk and Commenge [9]. Both reactor M1 and M2 found to be equivalent in performance, under some flow-rates, with certain micro structured reactors. For M2, at low dissipation rates around 10 W/kg, its mixing time performance corresponds with (sometimes much better than) that of single T-mixer with dimension of around 800 μm [11]. Mixing time of M1 is quite comparable, sometimes much better than, several micro designs like triangular interdigital micromixer by Mikroglas [11] (dimension 50 μm~150 μm), or Starlam by IMM [12] (dimension ~100 μm), etc.

It is also interesting to compare the performance between the two prototypes M1 and M2. By reducing the size of reactor by a half, the performance of mixing augments significantly. If properly handled, mixing time could be 4-5 times shorter for reactor M1 with the same energy dissipation than M2. Rapid mixing less than 4 ms can be realized using M1 when flow-rate is higher than about 400 mL/min for each fluid, showing very good micromixing performance.

Moreover, the throughput of both of them can be several hundreds of milliliter per minute, which is sometimes difficult for those micro structures to achieve. Our mini-structured
design makes the scale-up by numbering-up attractive. Simple design and fabrication may be easier for applications.

![Figure 9](image)

**Figure 9** Comparison of micromixing time with different studies

**CONCLUSION**

A high throughput mixer reactor with arborescent structure was designed and tested. Flow distribution uniformity is investigated by CFD simulation and optical tracer visualization. Micromixing performance was tested by using iodide/iodate method. Mixing time was estimated using IEM method with obtained segregation index.

CFD results show that in laminar flow, this structure will have good distributing feature between channels ($D_{ch}$ within $\pm 1.5\%$ under laminar flow conditions). The optical tracer visualization by fast camera also confirms qualitatively good distribution uniformity between channels, which is an essential feature for parallel channels application with the aim of numbering-up.

From the mixing characterization we found our millimeter design comparable to some micrometer products of IMM, Mikroglas, etc. This confirms the fact that we are not always obliged to go to micro scale structure only for obtaining a better mixing performance [13]. Millimeter scale (sometimes called meso scale) structures, if properly designed, could have good mixing performance without increasing much dissipation.

Although concerning surface-to-volume ratio, catalytic reaction or heat exchange do benefit more from micro channel design.

One drawback of this design is that the mixing is not enhanced in smooth straight channels. No split-and-recombine or turbulence enhances the fluid interaction during the flow. Further works will be done to decrease the distance of mixing inside channels by better design of T-mixers or by modifying channels structures. Modifying the walls of the inner channels using micro or mini-scale fins to enhance the mixing would be one interesting direction.

**REFERENCES**


