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# Recent progresses in intensified heat exchanger reactors with millimetric zigzag channels

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## **1. Context of the research activities**

Process intensification consists of designing or improving processes by reducing unit sizes. The origin of this concept dates from the 1980s, with the appearance of fixed rotating beds as an alternative to traditional packed columns for distillation [1]. Unlike the concept of "green chemistry" which emerged in the 1990s to help reduce pollution related to industrial chemical activities [2], the first objective of process intensification was to increase the profitability of factories. Indeed, reducing the size of units should in particular allow a reduction in investment costs, overall energy consumption and in the quantities of waste generated. This concept was then generalized to any improvement of a process making it possible to significantly reduce the size of the installation, its energy consumption, its environmental impact and to improve its intrinsic safety [3]. Process intensification is a major topic of research at The Chemical Engineering Laboratory of Toulouse (LGC UMR 5503) in France. In particular, the Science and Technology of Intensified Processes (STPI) group is interested in different ways of intensification for reaction, mixing and separation: compact reactors [4–6], photochemical reactors [7,8], supercritical CO<sub>2</sub> processes [9,10], innovative technologies for gas-liquid contactors [11,12].

For exothermic and fast reactions, continuous compact heat exchanger reactors offer exceptional thermal performance and constitute an interesting alternative to batch or fed-batch reactors still commonly used in fine chemicals for carrying out potentially dangerous reactions. The objective of intensifying processes is to favor thermodynamics and chemistry [13] by choosing the appropriate technology based on its performance in terms of mass and heat transfer rather than slowing down chemistry, particularly by dilution or by the use of fed-batch, to adapt to technological constraints. With this objective, the LGC and the CEA (Commissariat à l'Énergie Atomique et aux énergies alternatives, France) have collaborated in the 2000s to develop a heat exchanger-reactor based on plate technology in which millimetric zigzag channels of square section are engraved. The heat exchanger reactor is made up of an alternation of plates called "process" where a reactive medium circulates and called "utility" where a heating or cooling fluid circulates for thermal control of the device (Figure 1). The design proposed by LGC and CEA for the process channel consists in a zigzag millimetric channel that presents an edge of 2 mm engraved in a plate made out of a conductive material [14].

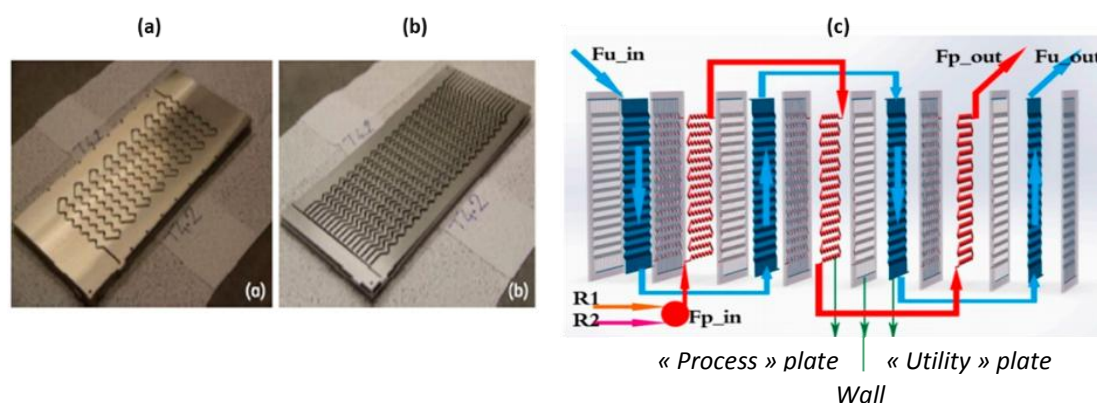


Figure 1. Configuration of the heat exchanger reactor designed by the LGC and the CEA and built by the CEA in stainless steel: (a) process plate, (b) utility plate composed of 16 millimetric zigzag channels in parallel and (c) assembly with an illustration of the possible fluid circulation (R1 and R2 would be reactants)

CEA built a device all made in stainless steel. A collaboration with the company Boostec Mersen allows the development of a HEX reactor entirely composed of silicon carbide (SiC) plates or as an assembly of SiC plates for the process plates and aluminum or stainless steel for the utility plates. In these devices, fast and exothermic reactions have been carried out such as fluorination [15], nitration [16] and hydrosilylation. The design for the heating/cooling fluid circulation in the utility plates may vary from one device to another but the process channel consists in a zigzag channel that presents a square cross-section and a design unit composed of a straight section 7 mm long and a 90° bend with a central radius of curvature of 1.5 mm. This channel presents a specific area of 2000 m<sup>2</sup>/m<sup>3</sup> and high heat transfer coefficient (around 3000 W.m<sup>-2</sup>.K<sup>-1</sup> for Reynolds number  $Re = 300$  and Prandtl number  $Pr = 4$  ; around 1200 W.m<sup>-2</sup>.K<sup>-1</sup> for  $Re = 16$  and  $Pr = 115$ ) [17]. The zigzag geometry generates flows tangential to the main direction of the flow so-called Dean's vortices [18], even at low Reynolds, that allows this good thermal performance with a moderate pressure drop penalty [19]. Moreover, the millimetric zigzag channel generates plug flow behaviour (axial dispersion coefficient of 3e-04 m<sup>2</sup>.s<sup>-1</sup> for an average water velocity of 0.1 m.s<sup>-1</sup> which gives a Péclet number  $Pe = 300$  for a reactor length of 1 m) [20] and high mixing time (around 1 s at  $Re = 200$ ) [21] which are essential to carry out fast reactions.

## 2. Recent progresses

Although the performance of the reactor with millimetric zigzag channels is no longer in doubt, this design is to this day often used to develop and test methods of characterisation useful for compact reactors, particularly to study temperature fields within the fluid in such devices. Indeed, the acquisition of local temperature fields in compact reactors is essential to understand the heat transfer mechanisms in these often-complex geometry devices, and to detect the presence of possible dead zones where hot or cold spots can appear during the implementation of reactions. In addition to poorly controlled conversion in these areas, they can lead to a significant deterioration in the quality of the final product and can be the site of dangerous reactions that can lead to thermal runaway of the reactor. The small dimensions of compact reactors do not allow the use of intrusive temperature measurement techniques (thermocouples or PT100) because they locally disrupt the flow. A non-intrusive optical thermometric technique often implemented is infrared measurement. However, it is not very accurate because it does not make it possible to obtain the temperature field directly in the fluid but measures the temperature of the wall in contact with the fluid by detecting the radiation from this wall. A spatially averaged value of the fluid temperature is therefore obtained. In addition, the measured field is diffuse due to the thermal conductivity of the

material. Finally, rapid temperature fluctuations cannot be measured due to the thermal inertia of the material. Thermometry based on Planar Laser Induced Fluorescence (PLIF), also non-intrusive, is a very good alternative because it has very good spatial and temporal resolution. This technique has been widely used for measuring temperature in the liquid phase (continuum, droplets and sprays) [22–24]. The so-called 2-colors/2-dyes method is recommended for its accuracy [24]. It requires the use of two fluorescent dyes whose temperature sensitivities in terms of fluorescence emission intensity are different and with distinct emission spectra. At LGC, the method has been implemented using fluorecein (FL, emission peak around 510 nm) and sulforhodamine B (SRB, emission peak around 590 nm) [25]. A pulsed Nd:YAG laser at 532 nm with a thickness of 500  $\mu\text{m}$  is used. As can be seen in Figure 2, two CCD cameras equipped with filters allow to mainly detect by one camera the fluorescence of FL, and by the other one the fluorescence of SRB. Heat-exchanger prototypes were built where the aqueous dye solution circulates in a square zigzag channel of 2, 3 or 4 mm wide, so-called “process channel”. Its inlet temperature can be varied between room temperature and 60°C. Water at room temperature circulates in a rectangular straight channel 12 mm wide and 8 mm deep, so-called “utility channel”. Both channels are engraved in PMMA plates and separated by an aluminum plate to allow heat transfer.

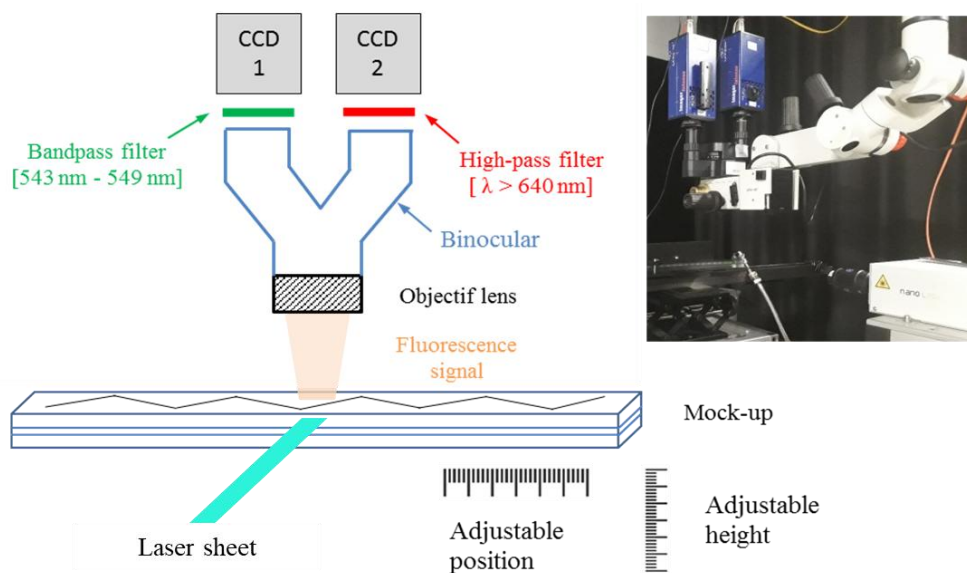


Figure 2. Experimental setup for the implementation of PLIF for temperature field measurement in millimetric zigzag channels

Figure 3 shows instantaneous images captured by both cameras. The field-of-view is slightly different due to small bias introduced by the filters. The temperature field over the common zone is determined thanks to a prior rigorous calibration of the method and a code developed in Matlab that notably uses its image processing toolbox.

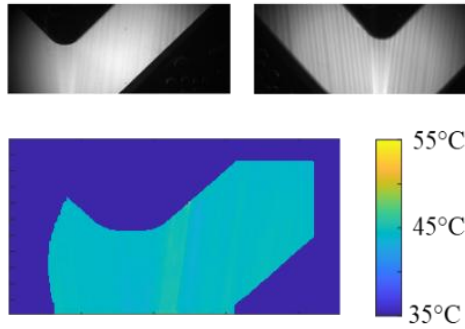


Figure 3. Images captured by both CCD cameras in the zigzag channel 3 mm wide and resulting temperature fields

The prototype is placed on a manual micrometric translation plate that allows the temperature fields to be measured at different depths and lengths in the zigzag channel. The obtained temperatures, averaged over the field-of-view, have been recently compared to numerical results in both laminar and turbulent flow regime (obtained from  $Re = 1000$  in the zigzag channels). The CFD software ANSYS CFX 2021 was used. Turbulent regime was computed with the SST model. Hydrodynamics and heat transfer are solved at the same time. The computational domain for the zigzag channel 3 mm wide is illustrated in Figure 4. It is composed of the process fluid channel and the aluminum plate. The exterior wall is practically in contact with the utility fluid. In the simulation, this wall temperature is supposed constant, equal to that of the utility fluid in the experiments which varies little from the inlet to the outlet of the utility channel. The position where the temperatures are compared are showed in Figure 4, corresponding to axial length of approximately 3 cm and 6 cm from the inlet.

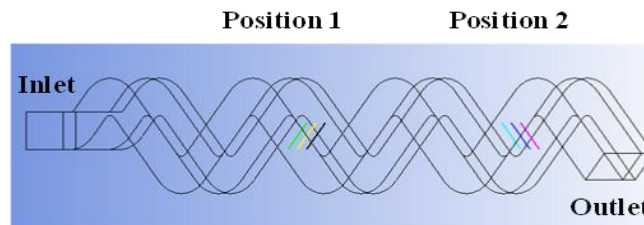


Figure 4. Computational domain composed of the fluid in the zigzag channel and the aluminum plate (at the right of the fluid)

The results obtained in the 3 mm wide channel according to the operating conditions are provided in

Table 1. Three planes in depth are investigated: high (2.25 mm above the aluminum wall),

	Laminar, $Re_{process} = 145$ , $T_{utility,in} = 31,6^{\circ}C$ $T_{process,in} = 47,9^{\circ}C$ , $T_{process,out} = 33,3^{\circ}C$				Turbulent, $Re_{process} = 1373$ , $T_{utility,in} = 40,5^{\circ}C$ $T_{process,in} = 56,4^{\circ}C$ , $T_{process,out} = 47,0^{\circ}C$			
	PLIF		CFD		PLIF		CFD	
Plane	Pos. 1	Pos. 2	Pos. 1	Pos. 2	Pos. 1	Pos. 2	Pos. 1	Pos. 2
High	45,1±0,6	43,5±0,6	47,3	46,2	54,6±0,6	54,5±0,3	55,3	55,1
Middle	44,9±1,0	43,7±0,9	43,2	42,5	55,1±0,2	54,5±0,3	55,0	54,8
Low	44,5±1,2	43,2±1,0	43,4	40,7	54,6±0,3	54,6±0,3	54,2	54,2

middle (at the center of the channel, i.e. 1.5 mm), low (0.75 mm above the aluminum wall). A decrease in temperature from the inlet to position 2 logically appears. The temperatures measured and obtained by CFD are in good agreement, in particular in turbulent flow where

the temperature fields are quite homogeneous over the channel cross-section. In laminar regime, CFD predicts larger temperature differences as a function of depth in the channel than experimentally. This can be due to the definition of the boundary conditions in the simulation, but also to the thickness of the laser sheet which reduces the spatial resolution of the observed fields compared to CFD.

Table 1. Temperatures (in °C) obtained experimentally by PLIF and numerically by CFD in a square millimetric zigzag

Plane	Laminar, $Re_{process} = 145$ , $T_{utility,in} = 31,6^{\circ}C$ $T_{process,in} = 47,9^{\circ}C$ , $T_{process,out} = 33,3^{\circ}C$				Turbulent, $Re_{process} = 1373$ , $T_{utility,in} = 40,5^{\circ}C$ $T_{process,in} = 56,4^{\circ}C$ , $T_{process,out} = 47,0^{\circ}C$			
	PLIF		CFD		PLIF		CFD	
	Pos. 1	Pos. 2	Pos. 1	Pos. 2	Pos. 1	Pos. 2	Pos. 1	Pos. 2
<b>High</b>	45,1±0,6	43,5±0,6	47,3	46,2	54,6±0,6	54,5±0,3	55,3	55,1
<b>Middle</b>	44,9±1,0	43,7±0,9	43,2	42,5	55,1±0,2	54,5±0,3	55,0	54,8
<b>Low</b>	44,5±1,2	43,2±1,0	43,4	40,7	54,6±0,3	54,6±0,3	54,2	54,2

channel 3 mm wide in laminar and turbulent regime

### 3. Future works

In future works, it would be interesting to take advantage of the recent developments in metal additive manufacturing to compare the performances of millimetric zigzag channels with square and circular cross-section. Indeed, additive manufacturing made it possible to build a stainless-steel exchange reactor designed by the LGC where two circular millimeter channels of 2 mm in diameter are intertwined inside a larger external tube which allows the circulation of a heating/cooling fluid. The printing was carried out by the company Materialise (France). The advantages of circular channel compared to square channel would be to maintain good heat transfer performance by reducing pressure drops and above all to reduce the phenomena of dead zones in the corners of the channel. Experiments need to be carried out to confirm these hypotheses.

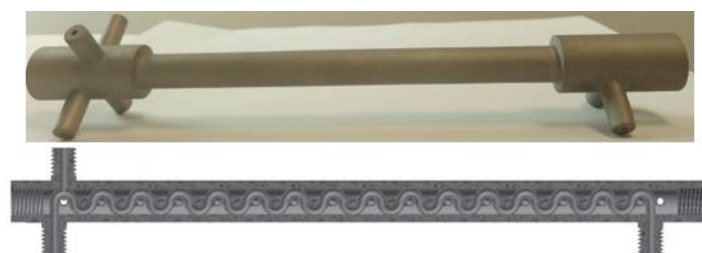


Figure 5. Heat exchanger reactor designed by LGC and build by metal additive manufacturing in stainless steel with circular millimetric meandering channels of 2 mm diameter

Also, to go further with the thermometry method by PLIF, it should be tested in other configurations of intensified reactors, for instance in complex liquid flows of film type. Thin films of a few hundred microns are notably obtained in Hi-Gee devices such as the spinning disc reactor. The variation in the film thickness from the center of the disk to the periphery and the wavy nature of the surface at high rotational speeds makes the measurement of

temperature fields by PLIF challenging. A combined measurement of the local film thickness and its temperature should be investigated.

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