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# 1D/3D Numerical Approach for Modeling a Two-Phase Flow in a Milli-Structured Heat Exchanger/Reactor

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Milli-structured heat exchangers/reactors are one of the intensified technologies, which have been successfully used in the case of exo- or endo-thermal chemical syntheses. Enhanced performances of such technologies compared to conventional apparatuses have been observed. This work is focused on the numerical investigation of the “DeanHex” heat exchanger/reactor with a 2D-meandering square cross-section channel with a hydraulic diameter of 2 mm. A new approach has been developed, using COMSOL Multiphysics V5.5, in order to model the reactor in a less resource demanding way than a full 3D approach. The  $2 \times 2 \text{ mm}^2$  meandering flow channel, which is the core of the heat exchanger/reactor design, is depicted by a 1D model coupled to a 3D model for the surrounding solid in which the channel is embedded. The interest of the 1D approach in modeling the milli-channel as a curve, lies in the significant reduction of the calculation time compared to 3D approaches, which are generally focused on local effects. It is a compromise between the simulation at small scale and the simulation at the reactor scale, giving a great advantage in computational efficiency over meshing and computing 3D channels. The 1D/3D approach is validated with heat transfer calculations between the 1D monophasic flow milli-channel and its 3D surrounding. The simulated results are compared to experimental ones, which were previously conducted on the “DeanHex” heat exchanger/reactor. The comparison shows a good agreement between temperature results. Small temperature differences (0.02–3 °C) are observed and are successfully explained. The next step is to integrate a liquid-liquid mass transfer model and to validate it with experimental results. The scale-up issue of “DeanHex” reactors will be then addressed, expecting a future industrialization of such equipment.

## 1. Introduction

Process intensification has been defined in the literature (Fernandez Rivas et al., 2020; Stankiewicz and Moulijn, 2000; Van Gerven and Stankiewicz, 2009) as a multidisciplinary engineering approach with the aim of developing innovative technologies, such as compact heat exchangers/reactors (HEX reactors), that offer more energy-efficient and environmental-friendly processes, with higher productivity and safer operation. Different kinds of HEX reactors exist and are summarized in the review of Anxionnaz et al. (2008). They are promising for the implementation of chemical syntheses, especially fast and exothermic ones, by offering a better heat transfer and temperature control. However, carrying out chemical reactions requires a sufficient residence time for the chemistry (laminar regime) and an efficient mass transfer. Therefore, corrugated geometries are implemented in compact HEX reactors. The plate HEX reactor (DeanHex) built by the Laboratoire de Génie Chimique (LGC, France) and the Commissariat à l'Énergie Atomique et aux Énergies Alternatives (CEA, France) is a fine example of such devices. It consists of a succession of process plates (where the reaction takes place) and utility plates (for cooling or heating), where millimetric channels are engraved by laser machining. In this work, a square process channel of a hydraulic diameter  $d_h$  equal to 2 mm with periodic zigzag units is considered. The geometry of the channel is represented in Figure 1. The radius of curvature of the bends  $R_c$ , the length of the straight section  $L_s$  and the angle between two straight sections  $\theta$  are equal to 1.5 mm, 7 mm and 90 °C, respectively.

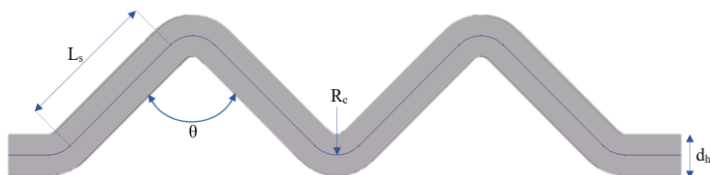


Figure 1: Schematic of the zigzag channel with geometric parameters.

The interest of such devices from a thermo-hydraulic point of view has been widely demonstrated in previous years (Anxionnaz-Minvielle et al., 2013; Théron et al., 2014; Tochon et al., 2010) and highly exothermic reactions were carried out successfully in the DeanHex without thermal runaway (Anxionnaz, 2009; Despènes et al., 2012; Di Miceli Raimondi et al., 2015; Elgue et al., 2012; Theron et al., 2012). Numerically, Moreau et al. (2017) built a 3D model for the DeanHex channels, using COMSOL Multiphysics 4.3b, allowing the estimation of axial dispersion coefficients from calculated velocity and concentration fields. In addition, a recent study has been carried out by Shi et al. (2019) aiming at characterizing the flow and heat transfer in the DeanHex square zigzag channel in laminar flow conditions. 3D simulations were solved using the finite volume CFD code, ANSYS CFX 16. However, in the 3D numerical approaches, a full 3D meshing of the wavy channel has to be constructed finely because of the bends. This leads to highly time consuming simulations. Therefore, to simplify the modelling of such channels, this work proposes a new 1D/3D numerical approach. The full description of this approach is detailed in this paper as well as its validation by previous experimental works conducted in the DeanHex millimetric zigzag channel.

## 2. 1D/3D numerical approach description

A full 3D numerical approach allows the direct modelling of local transfer mechanisms in the HEX reactor. However, given the complex geometry of the zigzag channel and the high  $L/d_h$  ratio, the 3D simulations are very costly in resources, especially in computing time. Therefore, in order to model the HEX reactor in a less resource demanding way, a new 1D/3D approach has been developed, using COMSOL Multiphysics V5.5. A 1D model is built for the meandering flow channel, using COMSOL's Pipe Flow Module. The latter solves for homogeneous cross sections, which means it performs averaged calculations in the channel's cross-sections. The modeled variables vary only along the length of the channel. Correlations, which were obtained during previous experimental works, are integrated in this 1D model. Consequently, this approach does not only help to avoid meshing the cross section of the channel with a full 3D mesh, but also offers a simulation method that is capable of predicting the performance of the reactor (reaction conversion, selectivity, etc.). The 3D channel is represented by a 1D curve "C" following its axial direction, embedded in a 3D surrounding solid "S". However, as seen from the solid matrix, the boundary condition between the channel and "S" is imposed by the channel on its axis "C" which is a line without any cross-section. In reality, this boundary condition is imposed on the walls of the channel with a  $2 \times 2 \text{ mm}^2$  cross-section, not on the axis. Therefore, in the present work, the cross-section of the millimetric zigzag channel, being non-negligible compared to the whole HEX reactor, must be taken into account in the model in order to correctly represent the DeanHex reactor. Consequently, the proposed solution is to define, around the curve "C", the three-dimensional channel "S2" as shown in Figure 2. Then a thermal chain "C-S2-S" is established to transmit the boundary condition from "C" to the walls of "S2". For that purpose, a radially "infinite" anisotropic thermal conductivity is applied to "S2" along with "negligible" density and heat capacity. A sensitivity study was carried out in order to determine the material properties of "S2" (density  $\rho$  ( $\text{kg} \cdot \text{m}^{-3}$ ), specific heat capacity  $C_p$  ( $\text{J} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$ ) and thermal conductivity  $\lambda$  ( $\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ )). The variations of the density and heat capacity of "S2" have showed no influence on the simulated thermal profiles. However, the effect of the radial thermal conductivity is clearly observed. As expected, by increasing  $\lambda$  to about  $1000 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ , the propagation of the temperature boundary condition from the channel "C" to the wall of "S2" is improved. Above this value, no more effect is observed on the thermal profiles. Consequently, to stay on the safe side, a radial thermal conductivity of  $10^5 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$  is applied to "S2" to transmit the "thermal information" from "C" to the walls of "S2", regardless of the system conditions.

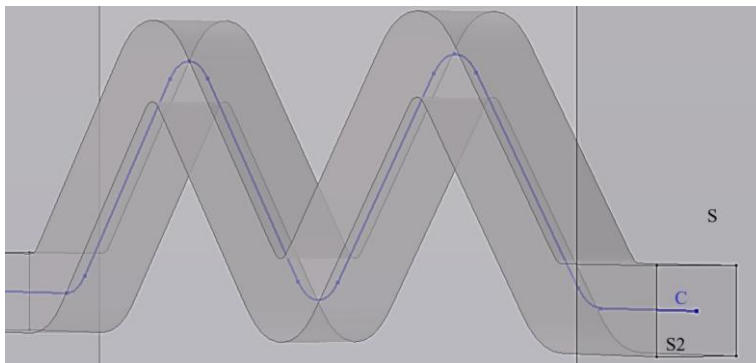


Figure 2: Illustration of the simulated geometry using the 1D/3D approach.

Since the geometry of the studied millimetric channel is composed of periodic zigzag units, imposing a radial infinite conductivity to "S2" requires the definition of a new curvilinear coordinate system (cc) for "S2". It allows the generation of a new base of vectors (ccx, ccy, ccz) at any point of "S2", as shown in Figure 3. The thermal conductivity of "S2" is thus defined according to this curvilinear coordinate system: infinite (equal to  $10^5 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ ) along ccy and ccz (green and blue vectors respectively) and zero along ccx (red vector).

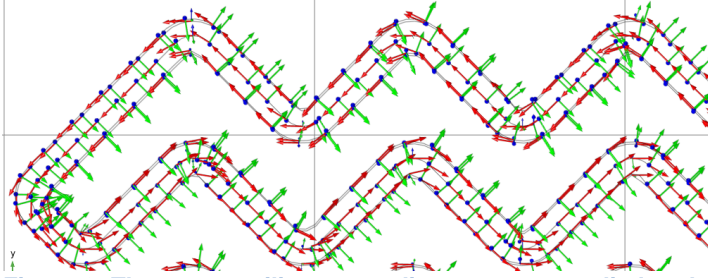


Figure 3: The new curvilinear coordinate system applied to the 3D zigzag channel.

### 3. Model equations

For incompressible fluid at steady state, the 1D pipe flow module calculates the pressure, velocity and temperature by solving the following momentum (Eq(1)), continuity (Eq(2)) and energy balance (Eq(3)) equations inside the channel referred to as the "process channel" (subscript p):

$$\rho_p u \square \nabla u = -\nabla p - f_D \frac{\rho_p}{2d_{h,p}} u |u| \quad (1)$$

$$\nabla \square (A_p \rho_p u) = 0 \quad (2)$$

$$\rho_p A_p C_{p,p} u \square \nabla T_p = \nabla \square A_p \lambda_p \nabla T_p + f_D \frac{\rho_p A_p}{2d_{h,p}} |u|^3 + Q_{wall} \quad (3)$$

$$Q_{wall} = h_p Z_p (T_s - T_p) \quad (4)$$

$u$  is the cross-section averaged velocity ( $\text{m} \cdot \text{s}^{-1}$ ),  $p$  the pressure (Pa),  $T_p$  the process fluid temperature (K),  $f_D$  the Darcy friction factor (dimensionless).  $f_D$  is correlated to Reynolds number  $Re_p$  for the millimetric zigzag channel as shown in Eq(5) (Théron et al., 2014). The process fluid physico-chemical properties such as  $\rho_p$  the density ( $\text{kg} \cdot \text{m}^{-3}$ ),  $C_{p,p}$  the heat capacity at constant pressure ( $\text{J} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$ ) and  $\lambda_p$  the thermal conductivity ( $\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ ) are assumed variable in function of  $T_p$ .  $A_p$  is the theoretical pipe cross-sectional area ( $\text{m}^2$ ) and  $d_{h,p}$  the mean hydraulic diameter (m) of the process channel.

$$f_D = 24.3 Re_p^{-0.71} \text{ for } 20 < Re_p < 200 \quad (5)$$

$$f_D = 6 Re_p^{-0.43} \text{ for } Re_p > 200$$

Furthermore,  $Q_{wall}$  is the heat exchanged ( $\text{W} \cdot \text{m}^{-1}$ ) across the channel's wall (Eq(4)),  $h_p$  the convective heat transfer coefficient inside the channel ( $\text{W} \cdot \text{m}^{-2} \cdot \text{K}^{-1}$ ),  $Z_p$  the wetted perimeter of the channel (m) and  $T_s$  the temperature (K) of the 3D surrounding solid. In this work,  $h_p$  is calculated from a Nusselt number correlation  $Nu_p$  (dimensionless) established for the DeanHex millimetric zigzag channel (Anxionnaz et al., 2011):

$$Nu_p = \frac{h_p d_{h,p}}{\lambda_p} = 0.16 Re_p^{0.66} Pr_p^{0.33} \quad (6)$$

Where  $Re_p$  and  $Pr_p$  are the Reynolds and Prandtl dimensionless numbers in the process channel, respectively. In addition, the resolved heat balance equation for the 3D surrounding solid "S" is given by:

$$\nabla \square (-\lambda_s \nabla T_s) = \nabla \square q_u \quad (7)$$

Where  $\lambda_s$  is the thermal conductivity ( $\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ ) of "S" and  $q_u$  the heat flux boundary condition ( $\text{W} \cdot \text{m}^{-2}$ ) imposed on the walls of "S". The latter will be further described in the following.

## 4. Experiments vs 1D/3D simulations

### 4.1 Description

The validation of the proposed 1D/3D approach is discussed by comparing the simulated results to experimental ones. The experimental work of Théron et al. (2014), in which thermo-hydraulic performances of the DeanHex millimetric zigzag channel had been characterized, is used. During this study, the HEX reactor consisted of a succession of three process plates sandwiched between four utility plates. 2 mm square cross-section channels, with periodic zigzag units, were engraved on both plates by laser machining. The process and utility streams are separated by a 2 mm metal thickness (closing plates). In the present work, only the first process plate is modeled (Figure 4a). The computational domains consist of a three-dimensional zigzag channel of total length equal to 2.2 m “S2” with a radially infinite thermal conductivity, centered by a one-dimensional zigzag curve “C”. “S2” is surrounded, from both sides, by a metallic solid matrix “S” of 2 mm thickness, representing the closing plates. The thermal chain “C-S2-S” is thus established as shown closely in Figure 4b. Nonetheless, the utility channels do not take part in the computational domains but are replaced by heat flux boundary conditions  $q_u$  at the upper and lower walls of “S”:

$$q_u = h_u(T_s - T_u) \quad (8)$$

Where  $T_u$  is the utility fluid temperature (K). The convective heat transfer coefficient  $h_u$  ( $W \cdot m^{-2} \cdot K^{-1}$ ) in the zigzag utility channels is deduced from the Nusselt correlation (Eq(9)) proposed by Shi (2019):

$$Nu_u = \frac{h_u d_{h,u}}{\lambda_u} = 0.2(Re_u^{0.67} + 8.9)Pr_u^{0.3} \left( \frac{d_{h,u}}{L_{s,u}} \right)^{0.4} \quad (9)$$

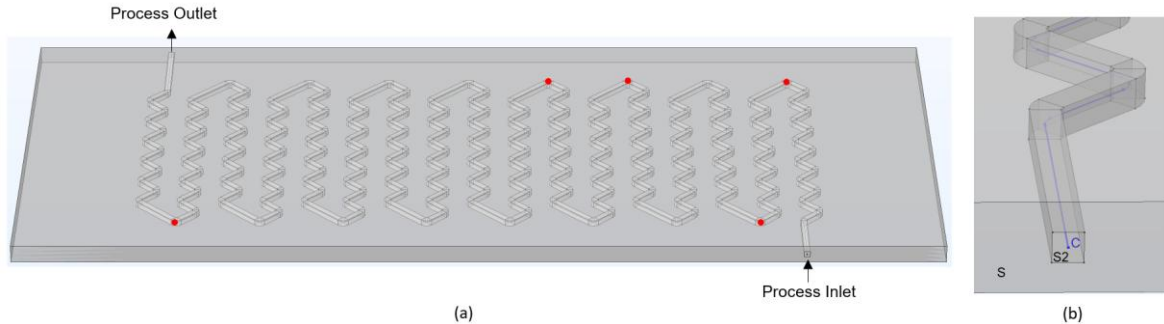
Where  $Re_u$  and  $Pr_u$  are the Reynolds and Prandtl dimensionless numbers for the utility side, respectively.  $d_{h,u}$  and  $L_{s,u}$  are, correspondingly, the hydraulic diameter (m) and the straight length (m) between the bends in the utility zigzag channel.  $\lambda_u$  is the thermal conductivity of the utility fluid (water) estimated at  $T_u$  as for all the other physicochemical properties of the utility fluid. A correction factor  $\frac{A_u}{A_s}$  is applied to  $q_u$  to consider the difference of heat transfer areas between the utility channels ( $A_u$ ,  $m^2$ ) and the walls of “S” ( $A_s$ ,  $m^2$ ).  $A_u$  is the heat transfer area of the utility channels which can be calculated using Eq(10):

$$A_u = P_u \cdot L_u \cdot N_u \quad (10)$$

Where  $P_u$  and  $L_u$  are correspondingly the perimeter (m) and the total developed length (m) of the utility channel.  $N_u$  is the number of utility channels engraved on one utility plate. Note that, the utility plate that is below the first process plate of the pilot, exchanges heat with the first and second process plates. Therefore,  $\frac{1}{2}P_u$  is used when imposing the heat flux boundary condition on the bottom side of “S”.

A free tetrahedral meshing is applied to the 3D domains where only thermal conduction occurs. Consequently, there is no need for a refined finely constructed meshing.

Furthermore, the experimental conditions are applied to the model (Théron et al., 2014). A constant temperature condition  $T_p$  is assumed (equal to about 75 °C) at the inlet of “C” and different water process flowrates  $Q_p$  ( $2.4 - 6.9 \text{ kg} \cdot \text{h}^{-1}$ ) are tested. Moreover, a constant temperature  $T_u$  is set (equal to about 15 °C) and the utility water flowrate  $Q_u$  ( $152 \text{ kg} \cdot \text{h}^{-1}$ ) was chosen in a way that guarantees that the heat transfer process is not limited by the local heat transfer coefficient on the utility side.



**Figure 4: Illustrations from the 1D/3D model of: (a) the process plate with the inlet and outlet of the flow (in red: the position of thermocouples in the experiments) and (b) a zoom-in on the process inlet where the S-S2-C chain is showed.**

## 4.2 Results

Since for the experiments the heat transfer process is not limited by the utility side, the comparison of the 1D/3D simulation results with experimental results will reveal any error that could be made on the process side and thus the proposed approach will be appropriately evaluated. In **Erreur ! Source du renvoi introuvable.**, the temperature evolution is represented in function of the axial position of the process channel, for different “process” flowrates. In the experiments, 5 K-type thermocouples were located along the first process channel, thus a part of the thermal profile is observed (Théron et al., 2014). As shown in **Erreur ! Source du renvoi introuvable.**, the simulated process thermal profiles are in good agreement with the experimental measurements. One aberrant point is observed and corresponds to the temperature measured at 0.63 m from the inlet. This discrepancy, observed for all the curves, can be explained by a misplacement of the thermocouple during the experiments. All the simulated temperature at this point are higher than that measured by this thermocouple. In fact, the fluid flowing through the process channel is normally hotter than the plate material. Therefore, the probe maybe in contact with the material of the plate instead of being inserted in the flowing fluid. Regardless this point, the maximum relative error is less than 8 %, which is assumed acceptable. Consequently, the 1D/3D approach with the radially infinite thermal conductivity imposed for “S2” is validated considering these experimental data.

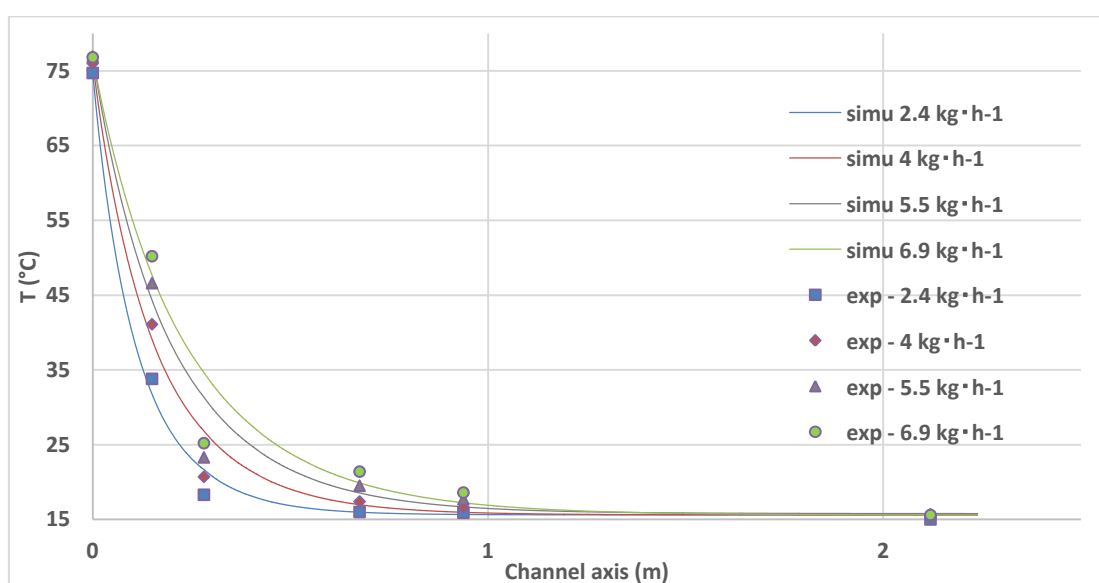


Figure 5: Evolution of RAPIC experimental and simulated process fluid temperature along the process channel for different process flowrates.

## 5. Conclusions

A new 1D/3D numerical approach is proposed in this work in order to model the DeanHex zigzag channel in a less resource demanding way. Avoiding the 3D meshing of the channel reduces significantly the computational time to about few minutes. By comparing the simulated results with experimental ones, the 1D/3D approach shows a great potential and seems promising. This leads to the the next step which consists of completing the heat transfer model by a mass transfer model and to validate it with experimental results. The present work is a first step towards addressing the scale-up issue of “DeanHex” reactors. Expecting a future industrialization of such equipment, a generalized scaled-up model will be used to predict the performance of the reactor in terms of conversion rate and energy efficiency at industrial scale.

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