Modelling of Heat Transfer in Microdroplets as Microreactors

Ferenc Ender¹, Gusztáv Hantos²
Budapest University of Technology and Economics
Department of Electron Devices
Budapest, Hungary
Email: ¹ender | ²hantos@eet.bme.hu

Abstract—A novel approach of modeling heat transfer in Taylor flow is presented. The model addresses especially the applications of Lab-On-a-Chip systems and focuses on the thermal behavior of individual droplets. Heat transfer is investigated at different boundary conditions as heat generation inside the droplet and heat generation at the droplet surfaces. A novel approximation of convective mass and heat transfer is presented as well.

I. INTRODUCTION

Segmented flows are attracting high attention due favorable heat transfer behavior in the microscale. It is widely recognised that such flows show a higher heat transfer coefficient practically exploited for cooling integrated circuits. Heat transfer issues were investigated by simulation by many authors [1]–[5]. Numerical models were presented as well by Muzychka et al. (2011), Walsh et al. (2010), Leung et al. (2010). [2], [3], [6]. Slug or Taylor flow as a special type of segmented flows has further advantages in the field of bioanalytics. This type of flow appears as consequent slugs of two phases which could be gas and liquid or two immiscible liquids and was first investigated by Taylor (1961) [7]. The aforementioned advantages are based on the following key features according to Theberge et al. (2010), [8]). The separated slugs form individual microreactors, which

- provide compartments in which species or reactions can be isolated
- are monodisperse and therefore potentially suitable for carrying out quantitative studies
- provide the possibility to work with extremely small volumes and single cells or molecules
- offer the ability to perform very large numbers of experiments

This paper addresses the bioanalytical use, where numerous investigation of possible applications and fluid mechanics including mass transfer were reported by many researchers, but the design aspects of thermal relations were not analyzed in depth. A review of the literature was presented by Theberge et al. [8]. As a general approach the thermal behavior of Taylor flow is usually investigated as a whole system comprising a few dozen of droplets in terms of cooling electronic devices. Though the numerous works are presented in this field numerical models are limited to average Nusselt number approximations which ignore the thermal relations of the individual droplets, and interprets all droplets together. In contrast, the thermal relations should be analyzed in individual droplets in case of monolithic microreactors as each droplet performs individual reaction. These reactions can occur on the droplet surface and/or inside the liquid slug and can vary in time as well, while the droplets, slugs and the channel wall itself are thermally coupled to each other. Therefore the
thermal modelling requires to count the droplets individually and the thermal couplings of the whole system as well.

Our final goal is to develop a thermal compact model which can be integrated in the design flow of monolithic microreactor based Lab-on-a-Chip devices. Thermal compact models are widely used in design practice of electrical systems, where these reduced order models yield fast results in early design phases contrary to finite element simulation which requires much more time. Typical simulation run-times of the Taylor-flow problem vary in wide scale from 38 to 1460 hours [4], [5].

A thermal compact model for integrated circuits with cooling microchannels was presented by Zhang et al. (2002), [9]. The model using thermal resistances and controlled voltage sources is able to predict the temperature distribution of the channel wall, but not for individual droplets. Numerical models based on Nu prediction were presented by Muzychka et al. (2011), Walsh et al. (2010), Leung et al. (2010). [2], [3], [6]. A compact model for modelling fluid dynamics in Taylor flow was presented by Jousse et al. (2005) [10].

This paper introduces a simplified model for modelling heat transfer in monolithic microreactors while keeping the running times between reasonable limits. The cases of surface and volumetric heat generation is detailed in depth while the classical constant heat flux boundary condition acts as a reference.

II. MODEL GENERATION

In order to generate the model the following requirements may be set. The model:

- is a good approximation of the heat transfer in Taylor flow at low Reynolds numbers typical for Lab-on-a-Chip systems,
- handles transient effects applied to the individual droplets,
- is linear, nonlinear parameters may also involved,
- involves any effects of internal forces (e.g. internal circulation) by simplified models and correlations,
- works with any types of boundary conditions (constant temperature and flux, heat generation inside the droplets etc).
- yields the local temperature variation and/or local heat flux coefficient as outcomes,
- describes the transient behavior of the system,
- is scalable. As a borderline case the model should be implemented by a special R-C network.

A. Setting up the model solver

As a first implementation of the model a finite element simulation in ANSYS Multiphysics 14 environment was performed. The model satisfying the aforementioned requirements was described in APDL (Ansys Parametric Design Language). The step-by-step process of the APDL code can be seen as a flowchart on Fig 1.

The two-dimensional geometry is defined first. Channel geometry, droplet size and length act as input parameters. Material and fluid parameters such as conductivity or heat capacitance, number of droplets, fluid velocity are also defined here. Any type of boundary conditions can be applied e.g. constant wall heat flux, constant wall temperature, volumetric heat generation inside the droplets etc.

A fixed frame computational domain is used which includes a part of the channel and a given number of droplets. In order to preserve the energy balance the number of droplets is kept constant during the simulation (the volumetric sum of the droplets is always constant).

The above steps are repeated until the desired simulation time is reached. Local values of the channel wall temperature and fluid mean temperature are available at all iteration steps.

The mesh is realized by a quadratic non-uniform meshing. The radial distribution of the nodes corresponds to the reverse Gaussian distribution, as the density of the nodes is higher at the wall-liquid interface region than in the middle of the channel. This non-uniformity of the mesh reduces the sum node number and therefore decreases simulation time. The distance of two node $d_n$ at the normalized axial position $r$ is

$$f(r, \mu, \sigma^2) = \frac{1}{\sigma \sqrt{2\pi}} e^{-\frac{1}{2} \left( \frac{r-\mu}{\sigma} \right)^2}$$

where $\mu = 3$ and $\sigma = 3$, the channel-diameter to channel-wall ratio is 10.2.

Material parameters are assigned to each node. Gas and liquid phase, channel wall and liquid film layers are treated as different materials.

Conduction and convection effects are considered by additional equations at affected nodes and will be discussed later.

A first, steady-state run is performed with all body force loads deactivated and the nodal temperature solution is saved. Mass
transfer is modelled by a movable mesh. The mesh of the fluid area is shifted by one node in each iteration step in the direction of fluid flow (Fig 2.). The nodes have a consecutive numbering from the left to the right and from the bottom to the top. Though the value of the nodes being shifted is formulated through a modulo function

\[ \text{shift}_{eff} = \text{mod(} \text{iteration}_n, \text{cellnum} \) \] (2)

where \( \text{iteration}_n \) is the number of the actual iteration while \( \text{cellnum} \) is the number of nodes in axial direction. Therefore the nodes behave as a closed loop, the droplet exiting at the outlet immediately enters at the inlet.

On the other hand the loop is opened from thermally point of view, as a Dirichlet boundary condition is applied at the inlet (2). The sum heat exiting at the outlet is applied as a Neumann boundary condition and calculated as follows:

\[ q_{\text{out}} = \frac{A \cdot \sum_{i=1}^{n} c_{\text{node}} \cdot T_{\text{node}}}{\Delta t} \] (3)

The nodal temperature result of the last run is loaded again as an initial condition in the following step. Body force loads are applied and a transient simulation is performed. Simulation time depends on fluid velocity as

\[ \Delta T = \frac{d_{n,ax}}{v} \] (4)

where \( d_{n,ax} \) and \( v \) are the axial distance of the nodes and the fluid mean velocity respectively.

The liquid film layer thickness is calculated on a basis of the correlation proposed by Aussillous and Quere states as follows:

\[ \delta_F = \frac{1.34Ca^{2/3}}{1 + 2.5(1.34Ca^{2/3})} \] (5)

where \( \delta_F \) and \( Ca \) is the film thickness and the capillary number, respectively. A mesh resolution of at least \( \delta_F/2 \) is set within the film area. The film layer is therefore modelled by a conductive liquid layer, as it was also suggested by He et al. (2009) [11].

B. Modelling convection

As a basic objective of the model it was stated that the solving time mainly caused by the iterative calculating of inertia forces must be reduced. In order to keep the model as simple as possible a constant velocity profile is used. This simplification is an essential condition to reduce the model complexity and therefore reduce running times. Obviously the approximation of the single phase parabolic velocity profile by a unit-step like function may cause an intolerable error. It will be shown that in Taylor flow the aforementioned errors may be satisfyingly handled.

- In laminar flow a thermal boundary layer develops very close to the wall and the characteristic effects of heat transfer occurs within this layer (assuming \( Pr \approx 1 \))
- In spherical bubbles the centerline part of the axial velocity profile more homogenous than in one phase flow thus the constant profile approximation does not result intolerable error.

- In thermal-entry length problems where the centerline part of the velocity profile is constant a higher Nusselt number is observed [12] in correlation with the experimental data observed by Talimi et al. [13]. In Taylor-flow a periodic thermal-entry length problem occurs. The thermal profile develops in every slug furthermore much faster than in single phase flow [5]. Therefore the sum error caused by the constant velocity profile approximation vanishes at the phase boundaries.

The aforementioned assumptions are reflected in Fig 5. where homogeneously mixed two phase flow under constant heat flux was simulated in Ansys FLUENT. The temperature rise from the channel inlet-to-outlet is plotted at different Reynolds number is observed [12] in correlation with the experimental data observed by Talimi et al. [13]. In Taylor-flow a periodic thermal-entry length problem occurs. The thermal profile develops in every slug furthermore much faster than in single phase flow [5]. Therefore the sum error caused by the constant velocity profile approximation vanishes at the phase boundaries.

![Fig. 4. Boundary conditions applied: a) constant wall heat flux, b) reaction at the liquid slug surface, c) reaction in the liquid slugs](image)

<table>
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<th>R value</th>
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...
C. Modelling internal circulation

The enhanced heat transfer experienced in Taylor flow is caused by the internal circulation of mass within the slugs. As the velocity field is not calculated by the model, the heat transfer enhancement effect of mass circulation should be considered by extending the conductive heat transfer equation by convective terms. Let us assume a slug with internal circulation and a constant heat flux applied at its upper tangent. As the upper part of the slug is heated, a thermal gradient occurs and heat is transferred in radial direction due to conductive heat transfer. Within the slug the heat is distributed by the constant mass circulation, therefore an improved heat transfer is experienced in radial direction, as it was experimentally shown by Muzychka et al. (2011) [6]. In axial direction thermal diffusion effects are dominant. The axial temperature distribution in the bubble changes rapidly due to about 20 times higher thermal diffusivity than of the fluid.

The above effects are estimated by a constant convective mass transfer applied at the different regions of the flow pattern regarding the dominant mass flow direction as depicted in Fig. 3. The mass velocity equals the fluid mean velocity thus the complete changing of the mass occurs within one slug-length as it was pointed out by Muzychka et al. [6]. The following equation is solved for each element (four nodes form an element)

\[
\int \left( \rho c \delta T \left( \frac{\partial T}{\partial t} + \upsilon \nabla T \right) + \nabla \cdot (\delta T) ([D] \nabla T) \right) = \int \delta T q^c dS_1 + \int \delta T h_f (T_w - T_m) dS_2 + \int \delta T q dV
\]

(6)

where \( \rho, c, q, [D] \) are fluid density, heat capacitance, heat flux and conductivity matrix respectively. Heat transfer occurs through internal fluid surfaces \( S_1 \) and fluid-wall interface \( S_2 \) at local wall and mean fluid temperatures \( T_w \) and \( T_m \). Heat is stored in the fluid volume \( V \), and finally the fluid may moving by mainstream velocity of \( \upsilon \). Note, that in axial direction the mass transfer is modelled by the moving mesh, and the second term of the first integral is only evaluated while modelling internal mass fluxes.

III. TEST CASES

Taylor flow of water and nitrogen gas with a capillary number of 0.0045 under different boundary condition has been solved. See material properties in Table I. A sum of 8 droplets were introduced into the channel of diameter 320\( \mu \)m and a mainstream velocity of 0.0028\( \text{m/s} \) to 0.00028\( \text{m/s} \) resulting a Reynolds number of 0.1 to 1. Channel wall is built of 30\( \mu \)m PDMS and a part of the channel wall is substituted by Silicon in order to modelling temperature sensor device. A constant wall heat flux boundary condition was used to validate the model as the analytical result of this case is known. Under constant heat flux boundary condition the surface heat flux is constant regardless which phase is present locally (Fig. 4a).

An approximation is used to model the heat generated due the surface reaction of the wall and the liquid slugs. This is the case where the wall is coated by enzymes and the liquid slugs carry their substrate. Regarding the Michaelis-Menten kinetics the enzyme reaction could constantly generate heat if the substrate concentration is much higher than the enzyme concentration. It could be proven by the mass circulation inside the liquid slugs, therefore the heat generation effect should be more intensive where the circulation effect is dominant. This function is approximated by the Fourier row of the unit step function as follows:

\[
q_{\text{sur}}(x) = h \cdot \left( 1 - (0.5 + 0.5 \cdot (\cos(x) + 0.5 \cos(x)^2)) \right) \quad (7)
\]

where \( h \) is the heat flux constant and \( x \) is the axial position (Fig. 4b).

Another relevant case is when the biological reaction occurs inside the liquid slugs (i.e. both the substrate and the catalyst are present inside the slug). This case is approximated a constant volumetric heat production for each node of the liquid volume (Fig. 4c).

The possibility of sensing the temperature changes develops due to biological reactions was also considered. A sensor chip made of silicon was inserted the channel wall just before the outlet. All heat flux generation were ignored at the sensor area. The thickness of the sensor chip is 30\( \mu \)m. The time function of the average temperature of the sensor chip was investigated. The problem was solved for different sensor sizes and liquid slug lengths. The liquid slug is 560\( \mu \)m (short slug) or 1120\( \mu \)m (long slug). According to the slug length in case of short slugs the sensor length is 400\( \mu \)m and 650\( \mu \)m for short and long sensors, respectively. In case of long slugs the sensor length is 800\( \mu \)m (see Table II).
IV. Results

A. Performance analysis

The model implemented in ANSYS Multiphysics 14 was run on an Intel Core i5 4 core CPU with 4 GB RAM under 64 bit Debian GNU/Linux (kernel 3.2.0-2-amd64). In order to achieve reasonable results an axial resolution of $80\mu m$ was applied for a channel diameter of $320\mu m$ which yields a sum of 2736 nodes. For the channel length of $6\text{mm}$ a sum of 75 iterations are needed to achieve the thermal equilibrium resulting a total runtime of 20 minutes.

The build up of the model requests 4.96s while the model solver runs 13.29s in each iteration. The solve-time-to-build-time-ratio is therefore 2.68 which grows rapidly for more complex problems, for a problem size of 21888 nodes the ratio becomes 3.94. This issue raises the need of parallel processing to reduce solution times. Figure 7 shows the build and solve times for different problem sizes and for serial and parallel processing. It can be seen that for smaller problem sizes the serial processing (1 CPU) gains shorter run times both for model building and solve processes. The 1 CPU-to-4-CPU-runtime-ratio is 0.77 for 2736 nodes, however even for a bigger problem of 21888 nodes it remains below one, exactly 0.91. The iterations could not be threaded as the result of the last run is always requested, parallel processing makes sense only at huge node numbers. It is expected that for more bigger problems the parallel processing gains shorter runtimes than the serial one.

B. Thermal profiles

Thermal profiles along the channel axis was investigated under various boundary conditions detailed before. The temperature profiles are plotted at the axial position of the $\frac{Z}{c} = -1.1$ at different boundary conditions (Fig 6.) It can be seen that the thermal profile can vary characteristically under different boundary conditions applied.

For constant wall heat flux boundary condition the usual solution for thin wall was experienced. Due to the very moderate heat capacitance of the wall and very low Reynolds numbers of $Re = 1$ the wall can reach the thermally stable state and the wall and fluid temperatures run parallel. As heat generation is present at both phases the wall-fluid temperature difference does not differ significantly along the channel.

In case of surface reaction modelling, heat generation only occurs at the liquid slug. The heat generation rate depends also on axial position based on Equation 7. The temperature rises rapidly from $Z_c = -1$ to $-0.5$ where the slope of the temperature rise decreases due to the temperature gradient between the slug and gas bubble. Regarding the significantly smaller heat capacitance of the gas bubble its temperature reaches one of the liquid slug. Due to conductive heat transfer from the wall its temperature could be slightly higher than the slug’s. Due to the high heat capacitance of the liquid slugs, far enough heat can be stored to drive the temperature sensor.

Temperature gradients develop another way in case of modelling volumetric heat generation. Obviously a much bigger amount of heat is generated under this boundary condition. The heat generated at the liquid-gas surface heats up the gas bubble while its mean temperature rises above the liquid slug’s temperature. Due to the internal heat generation the temperature is higher at the channel centerline then those of the wall, unlike all other cases. That is why the temperature of the gas droplet is higher than the wall temperature as well.

C. Temperature measurement analysis

The temperature of the liquid slugs is measured by a silicon sensor modelled by a silicon-plate inserted in the wall. Due the $1 : 33000$ ratio of heat conductance of PDMS and silicon the heat stored in the liquid slugs and gas bubbles is conducted rapidly into the sensor substrate and rises its temperature. The temperature rise becomes proportional to the amount of heat stored inside the phases. Note, that temperature sensing should...
be applied to the slugs individually, so each slug should be separated by the measurements. That is why the phases should be measured separately otherwise the heat differences of the phases would be averaged decreasing the sensor signal.

The main properties of the test cases can be seen in Table II. The results of the test cases are compared to each other through the amplitude of the output signal $T_{osc}$ (the time dependent mean temperature of the sensor) and a normalized value called $R$ and defined as follows:

$$R = \frac{T_{osc}}{T_{max}}$$  \hspace{1cm} (8)

where $T_{max}$ is the temperature of the droplet exiting at the outlet.

In the cases 1, 5 and 6 the sensor area is longer than the liquid slugs resulting the aforementioned averaging effects therefore gains very low $R$ values. Worse values do not affected by the way of heat generation. Low $R$ value may be also experienced if the ratio of slug and sensor heat capacitance is too small. In case 2 and 7 short slugs are flowing under surface heat generation which results very low oscillation and $R$ value as well.

Better results can be achieved by keeping the length of slugs higher than the sensor length. Case 3 and 4 show good $R$ values regardless the way of heat generated. Obviously higher oscillation amplitude can be achieved by volumetric heat generation in case 4.

Droplet velocity can also affect both $T_{osc}$ and $R$ values. Lower velocities enable longer contact time at the sensor yielding higher heat accumulation. Lower velocity means also less convective heat transfer ratio increasing the slope of the temperature profile along the channel, which results also higher temperature. Case 8 and 9 are reflecting the aforementioned facts yielding very reasonable $R$ values.

V. Conclusion

Based on the general properties of Taylor flow a simplified model was introduced to describe heat transfer in microchannels. In order to validate the model it was implemented in a finite element solver, ANSYS Multiphysics. Note the results of temperature measurement analysis where the same heat generation rate was applied in all cases. The effectiveness of temperature sensing can vary by a factor of 10 for the same heat generation rate only by changing the droplet size, velocity and the sensor size.

An other limiting factor of heat loss, due to heat convection or radiation from the channel outer surface was ignored in this analysis. This factor could worse the $R$ values at low velocities which forms the problem into an optimization task.

As all of the equations building up each finite element used in this model could be substituted linear electric circuit models, the model is suitable to be implemented as a linear network. In the future work the possibilities of such implementation will be investigated. Model parameters regarding the kinetics of biological reactions should be also refined by using more complex models.

REFERENCES


