Modèle à 2T

Pour la caractérisation thermique d’écoulements biphasiques en millifluidique

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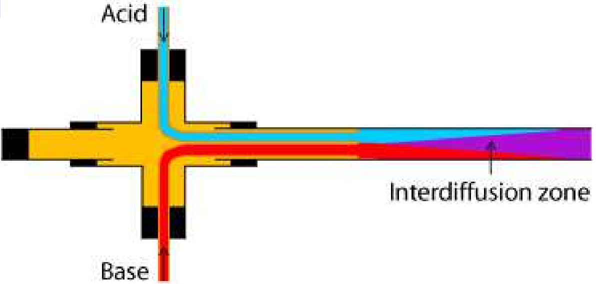
Goals: Determination of kinetic and enthalpy of chemical reaction or phase change in millifluidic droplet flow

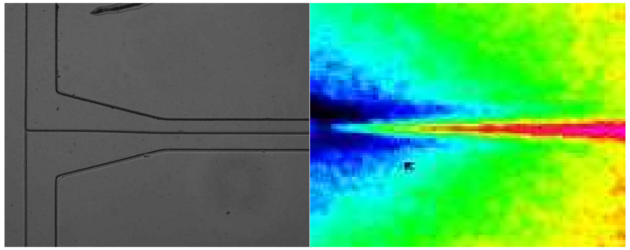
❶ Applications: Chemical reaction in co-flow or droplet

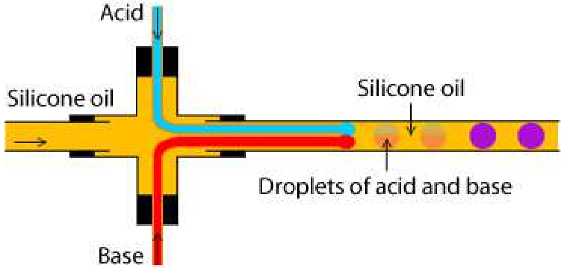


Laminar flow (low Reynolds number)

Mixing by species diffusion

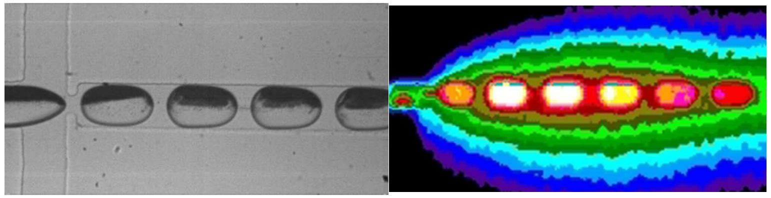
Steady state (x = vt)





Mixing is faster

1 droplet = 1 batch microreactor

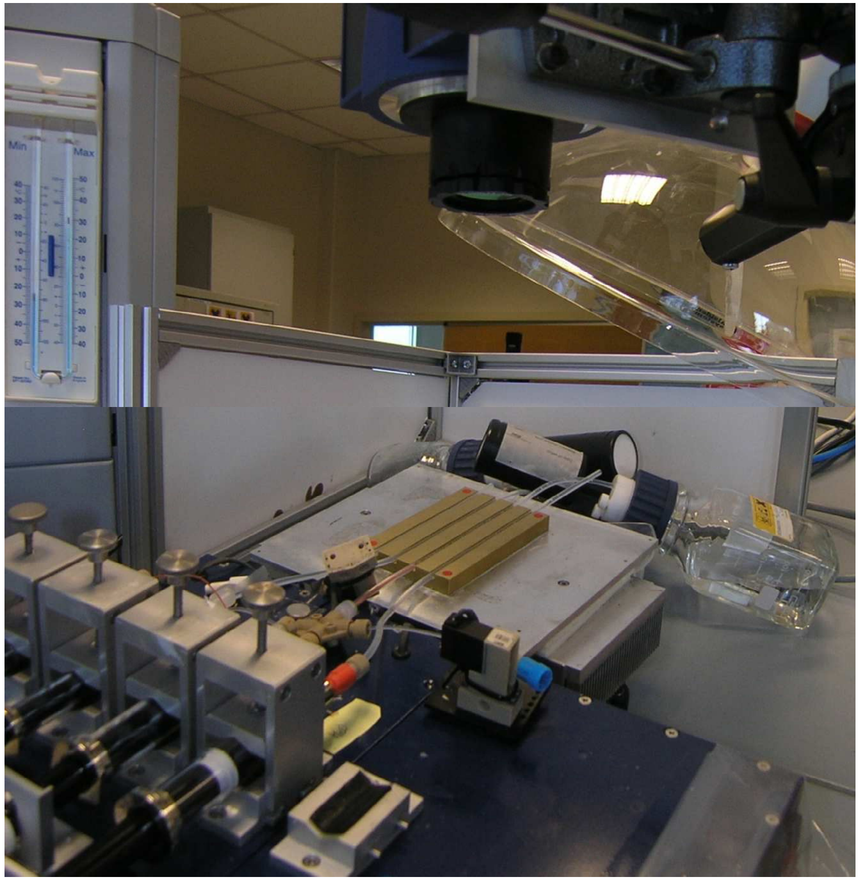


❷ Drawback : Estimation of ***fields of thermophysical properties*** (thermal properties, flow properties, heat sources) in ***fluidic chip*** during thermal process

$$div\left(\lambda . \overrightarrow{grad}\ T(x,y,t)\right) + \phi(x,y,t) = \rho C_p \left(\frac{\partial T(x,y,t)}{\partial t} + u \frac{\partial T(x,y,t)}{\partial x} + v \frac{\partial T(x,y,t)}{\partial y} \right)$$

❸ Tools : ***Temperature fields measurements at microscales*** (InfraRed camera), ***analytic and numerical modelisation*** (quadripole, finite difference…) and ***inverse methods*** (nodal: OLS, TLS and modal: SVD approaches)

Goals: *Determination of kinetic and enthalpy of chemical reaction or phase change in millifluidic droplet flow*



Advantages of millifluidic reactor

- Small volumes of reagents (µl)
- Study of chemical reactions under flow
- Enhancement of transfer phenomena
- Operates under isoperibolic thermal conditions (control of the temperature of the chemical reaction).

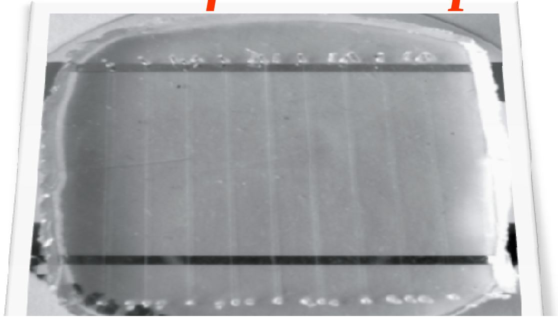
IR Thermography + Inverse Methods

- IRT technique is applied to measure the temperature profile
- Inverse method based on analytical simplified model is used in order to obtain thermokinetics parameters such as enthalpy

Spectroscopy

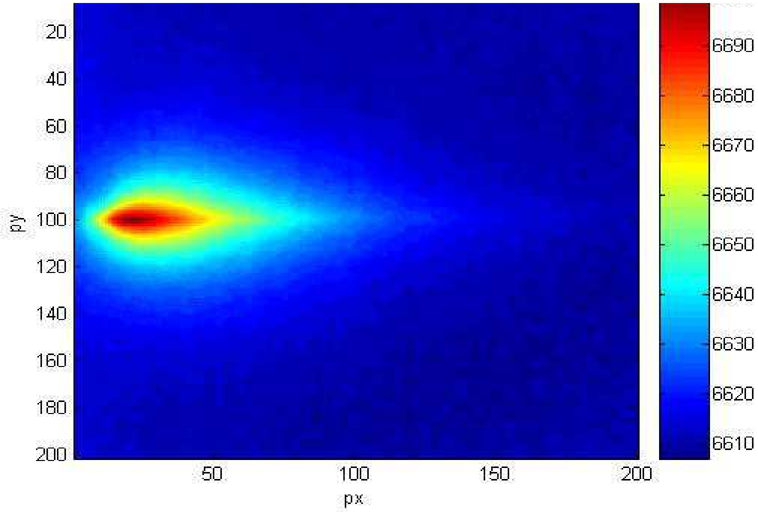
- Evolution of chemical reactions can be followed, in order to measure the experimental conversion by online analysis.
- Allows high accurate characterisations of the kinetic parameters of chemical reactions.

Microfluidic chip

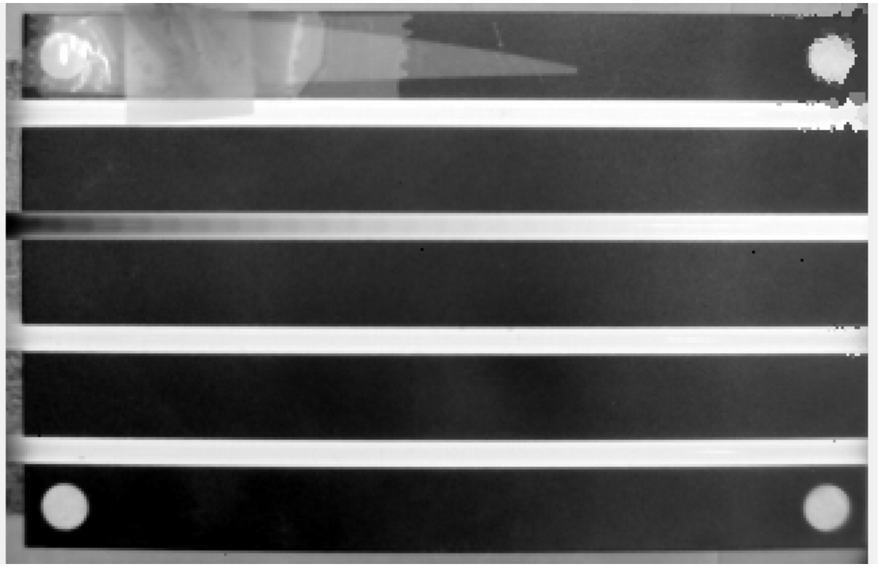


Millifluidic chip

IR temperature



IR temperature



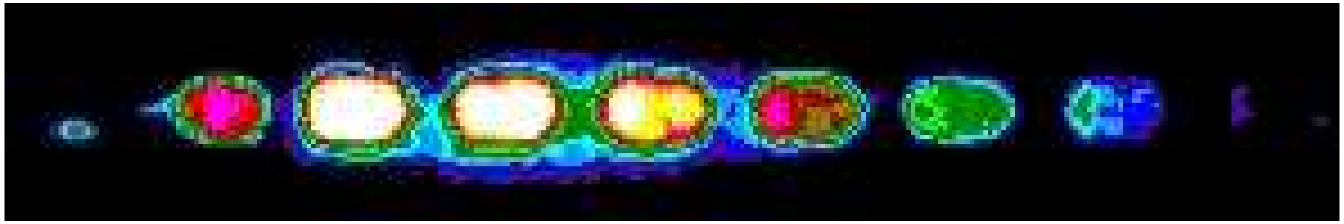
The thermal transfer is easier

No diffusion outside the channel

Well known boundary condition

Main drawback

How to estimate thermophysical parameters inside such complex system

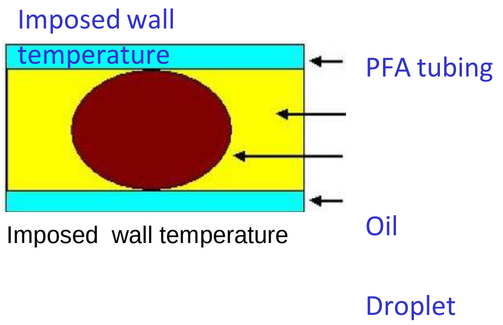


Direct thermal modelisation

$$div(\lambda.\overrightarrow{grad}\,T(x,y,t))+\phi(x,y,t)-hS\left(T(x,y,t)-T_0\right)=\rho C_p\left(\vec{v}\,\overrightarrow{grad}\,T(x,y,t)+\frac{\partial T(x,y,t)}{\partial t}\right)$$

Heavy inverse data processing

Diffusion in biphasic media (water-oil) + wall
Convection between droplets and oil
Chemical reaction into the droplet
Field of velocity

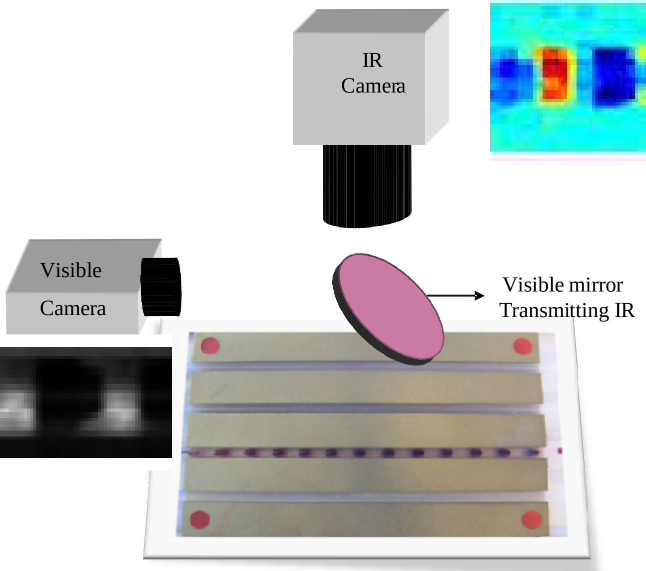


Simplified the thermal study

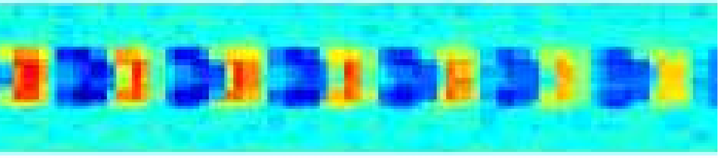
Working in the space of the droplet
Neglect the diffusion along x direction (S/V, Bi<1)

First idea: simplified the thermal conditions

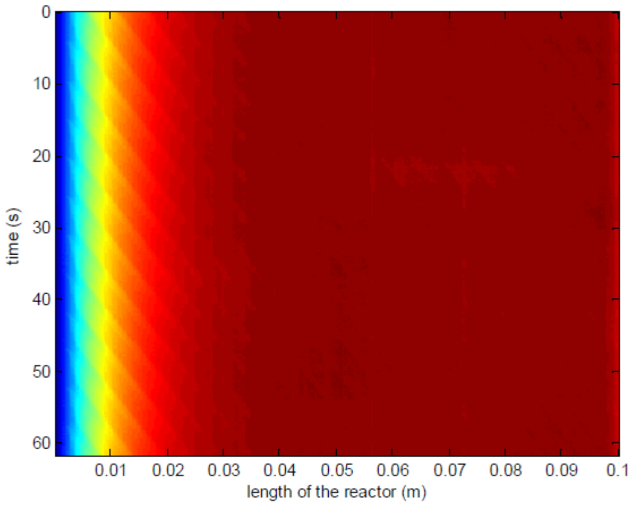
Working directly into the droplet space (Lagrangian transformation)

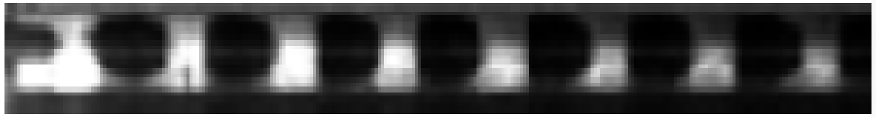


Experimental device

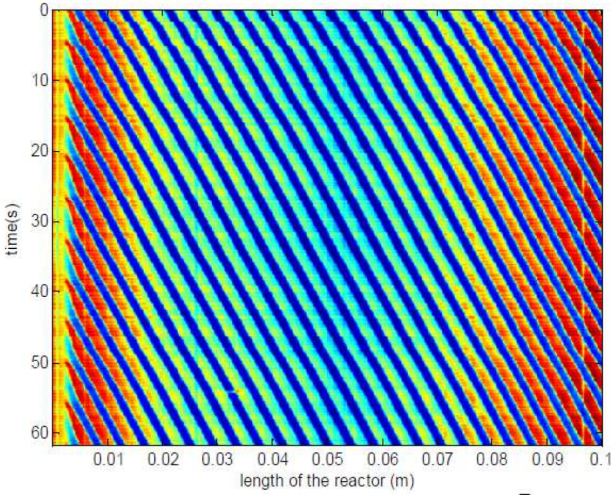


Measured temperature





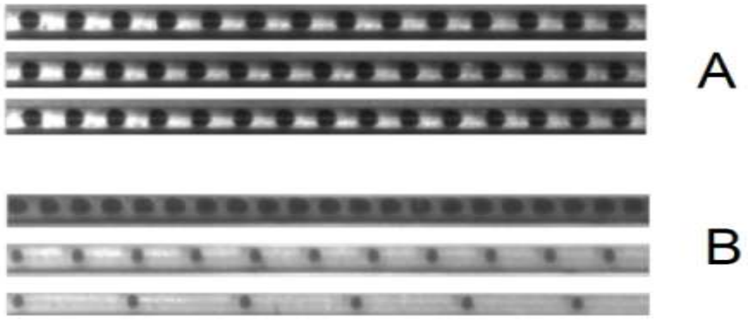
Measured visible droplets

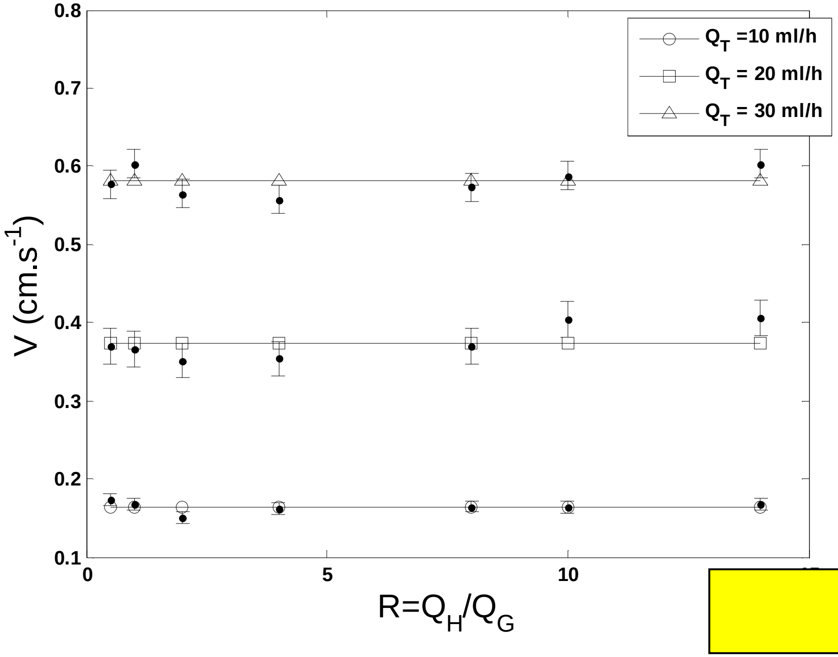


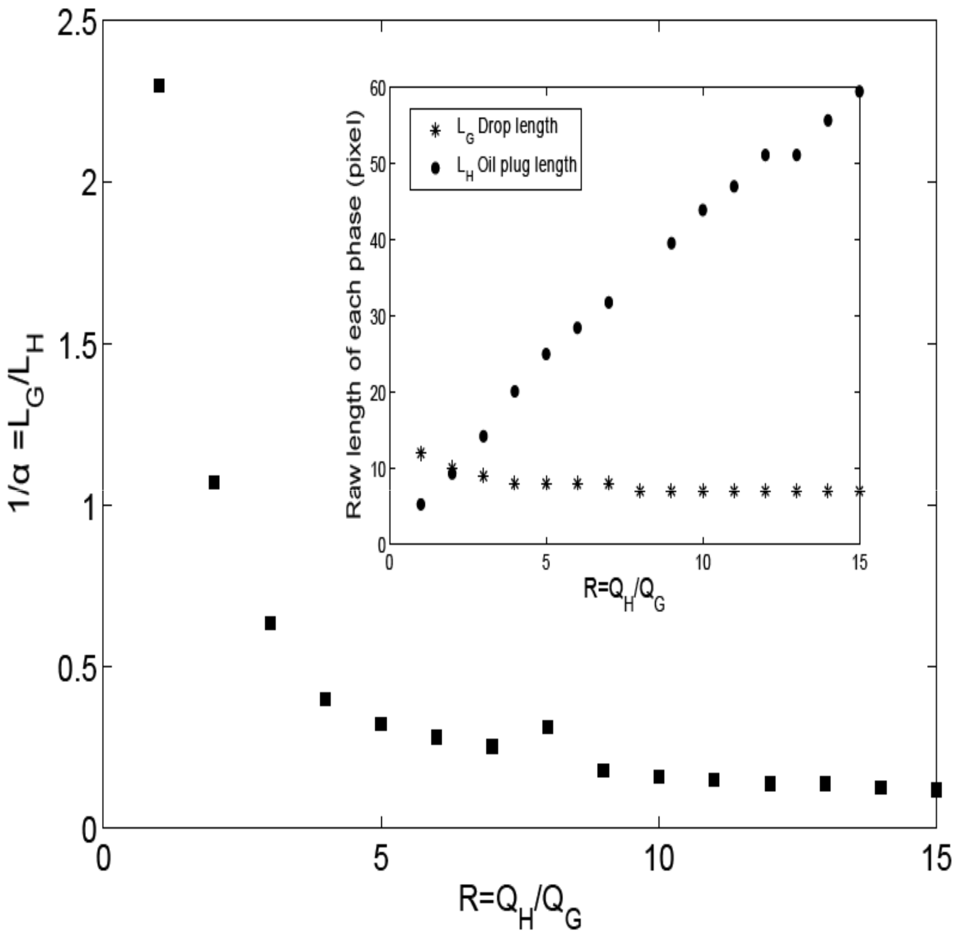
Estimation of the mean velocity
Evolution versus time of droplet and oil temperatures

First idea: simplified the thermal conditions

Working directly into the droplet space (Lagrangian transformation)

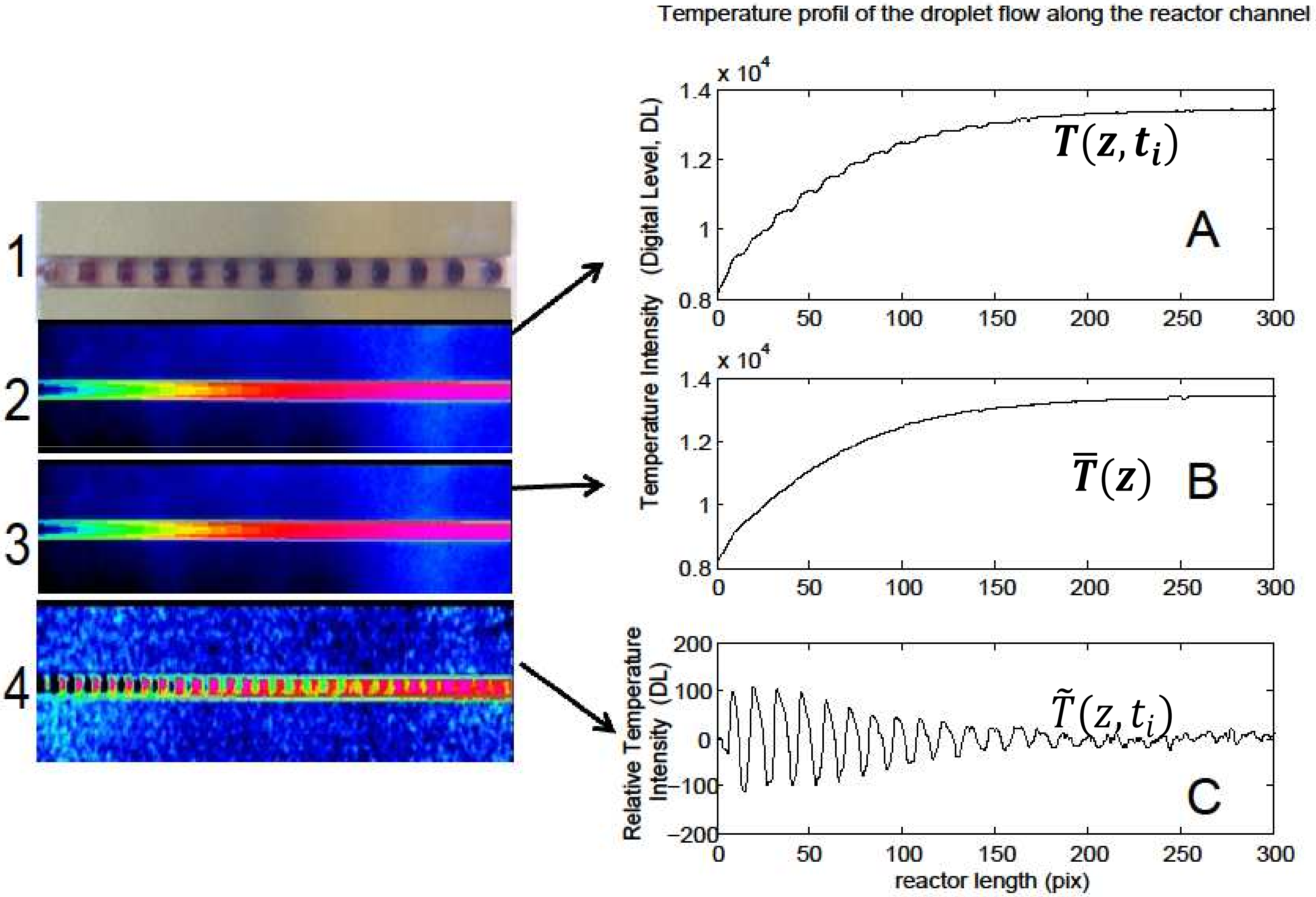






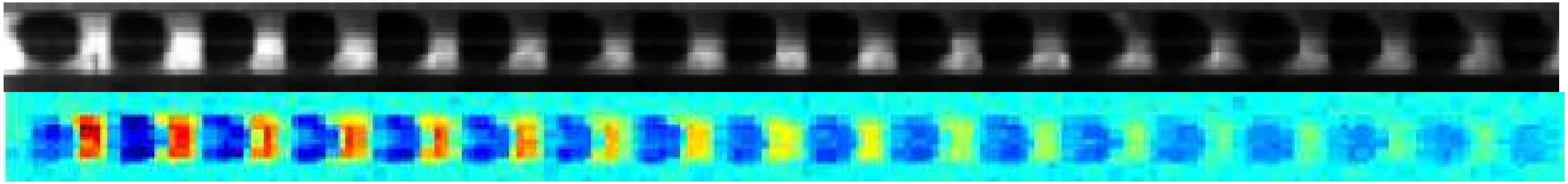
Very good hydrodynamic stability

Second idea: Assuming the problem can be seen as a periodic established one

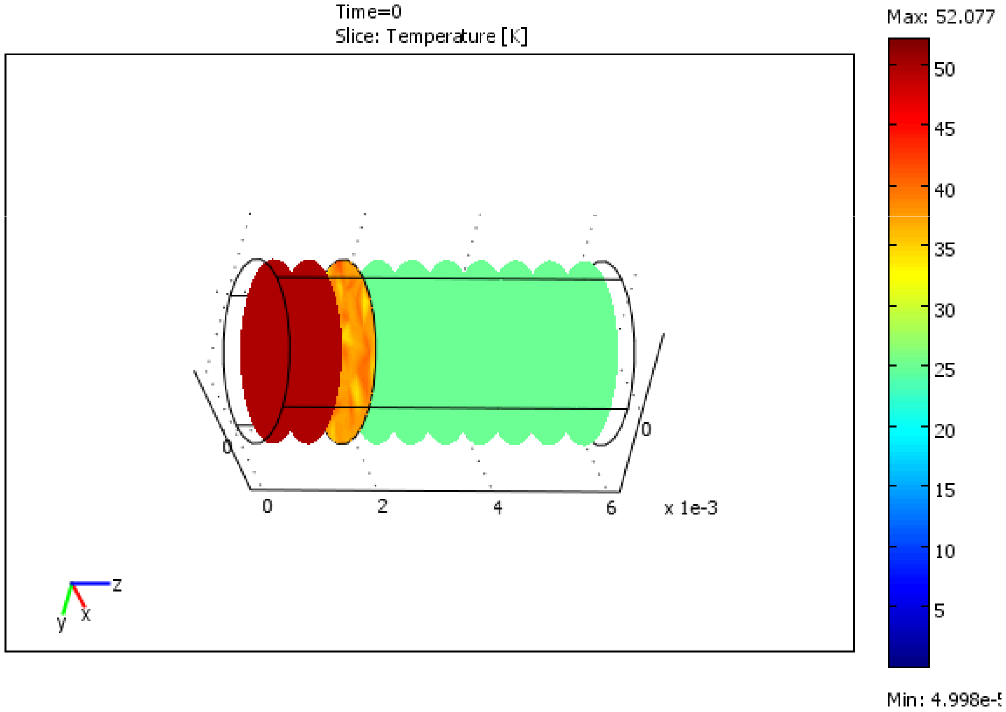


Third idea: Neglect the thermal diffusion between the two phases

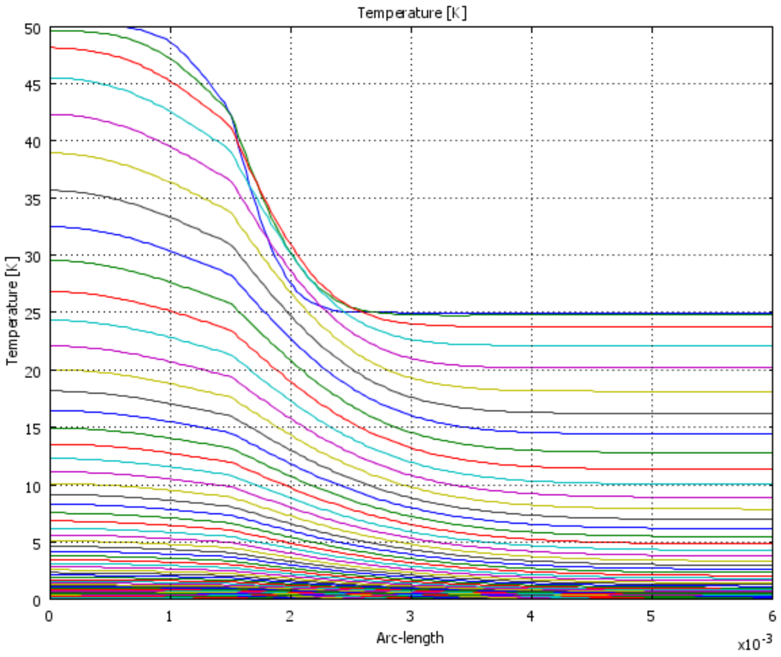
Visible and IR images



Analytical simulation in the space of the plugs

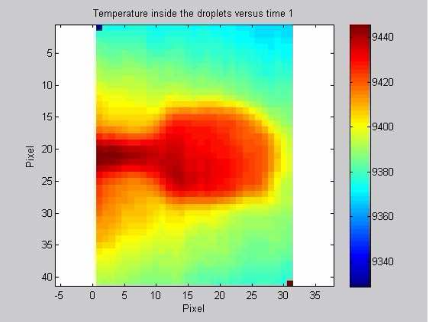


Analytical temperature profile along z as function of time



Question: Use a thin body approximation (neglect z diffusion)

Used of thin body thermal model



Averaging of the temperature

$$\frac{dT_G}{dt} = \Phi - H_1\left(T_G - T_P\right) - H_2\left(T_G - T_H\right)$$

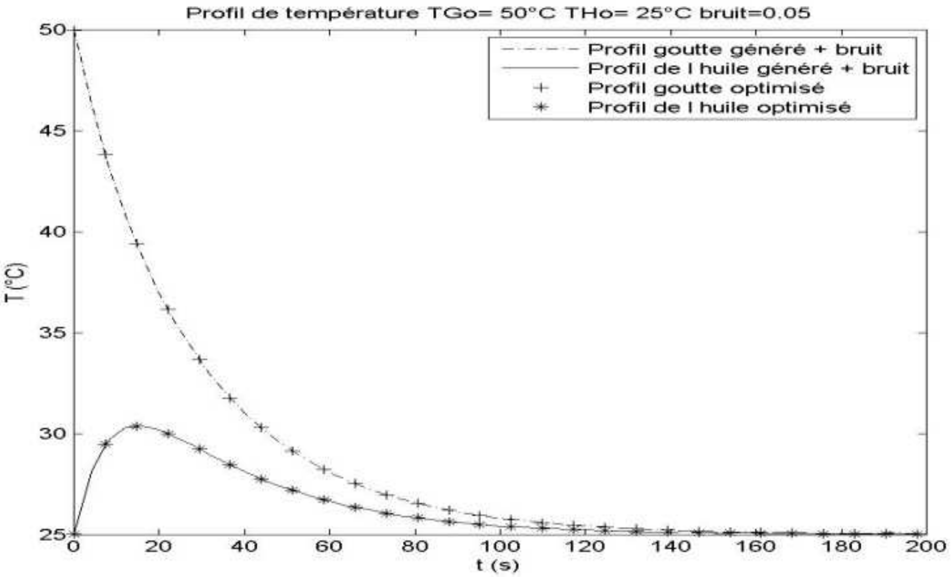
$$\frac{dT_H}{dt} = -H_3\left(T_H - T_P\right) - H_{2p}\left(T_H - T_G\right)$$

$$H_1 = \frac{h_{GP}S_{GP}}{\rho Cp_G V_G}; \quad H_2 = \frac{2h_{GH}S_{GH}}{\rho Cp_G V_G}; \quad H_3 = \frac{h_{HP}S_{HP}}{\rho Cp_H V_H}; \quad H_{2p} = \frac{2h_{HG}S_{HG}}{\rho Cp_H V_H}; \quad \Phi = \frac{\phi}{\rho Cp_G V_G}$$

Resolution in Laplace domain

$$\theta_G = \frac{\left(H_1\frac{\theta_p}{p} + T_p\right)\left(p + H_3 + H_{2p}\right) + \left(H_3\frac{\theta_p}{p} + T_p\right)H_2}{\left(p + H_1 + H_2\right)\left(p + H_3 + H_{2p}\right) - H_2 H_{2p}}$$

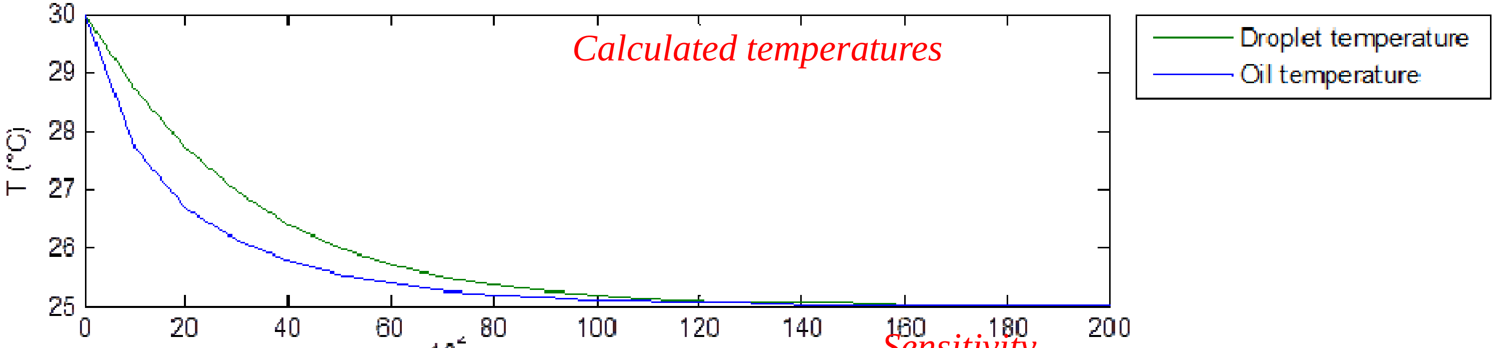
$$\theta_H = \frac{\left(H_3\frac{\theta_p}{p} + T_p\right)\left(p + H_1 + H_2\right) + \left(H_1\frac{\theta_p}{p} + T_p\right)H_{2p}}{\left(p + H_1 + H_2\right)\left(p + H_3 + H_{2p}\right) - H_2 H_{2p}}$$



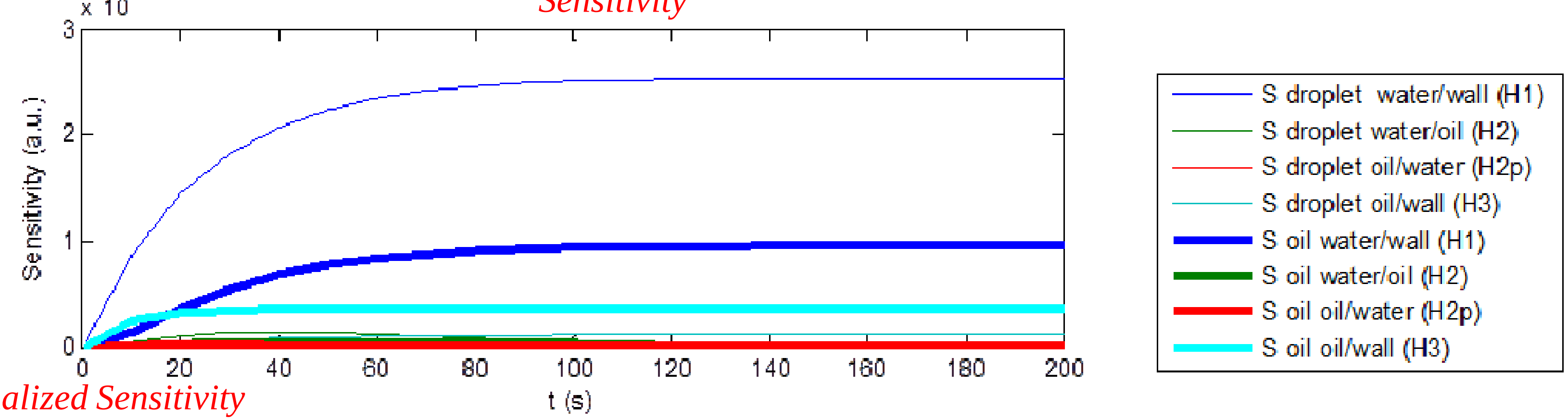
Drawback: Estimation of the heat exchange coefficients

Sensitivity study $S_\beta = \frac{\partial T}{\partial \beta} \times \frac{1}{\beta}$

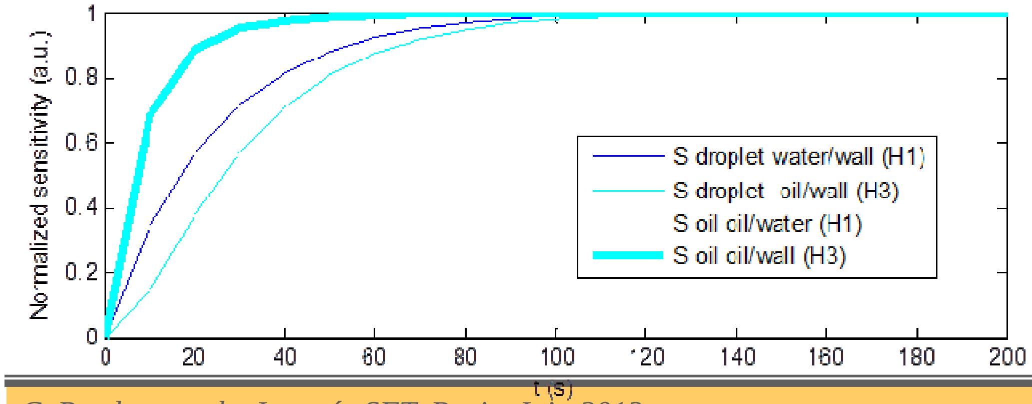
Fluids at T0=30°C and plate at Tp=25°C


Calculated temperatures

Sensitivity



Normalized Sensitivity

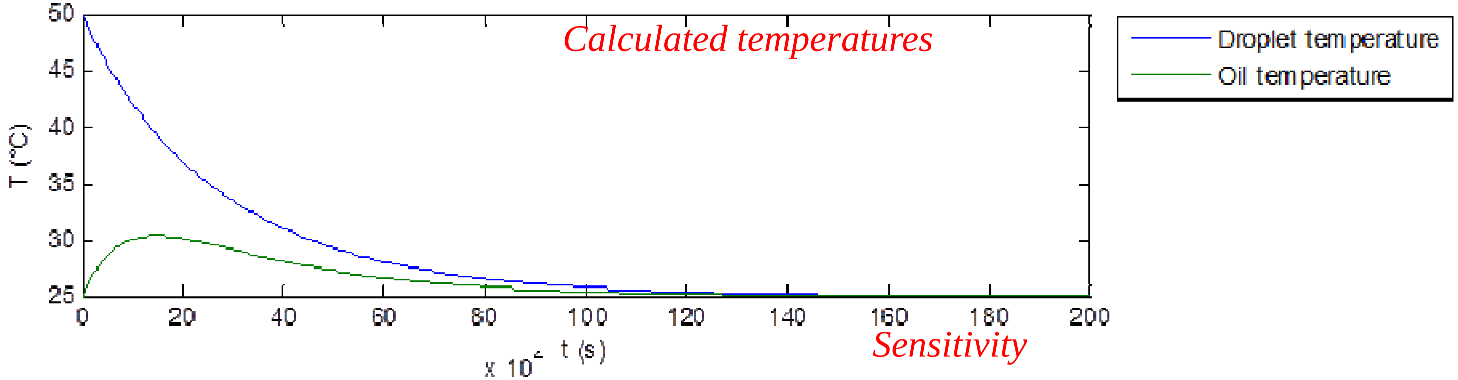


High sensitivity to heat exchange between fluid and plate

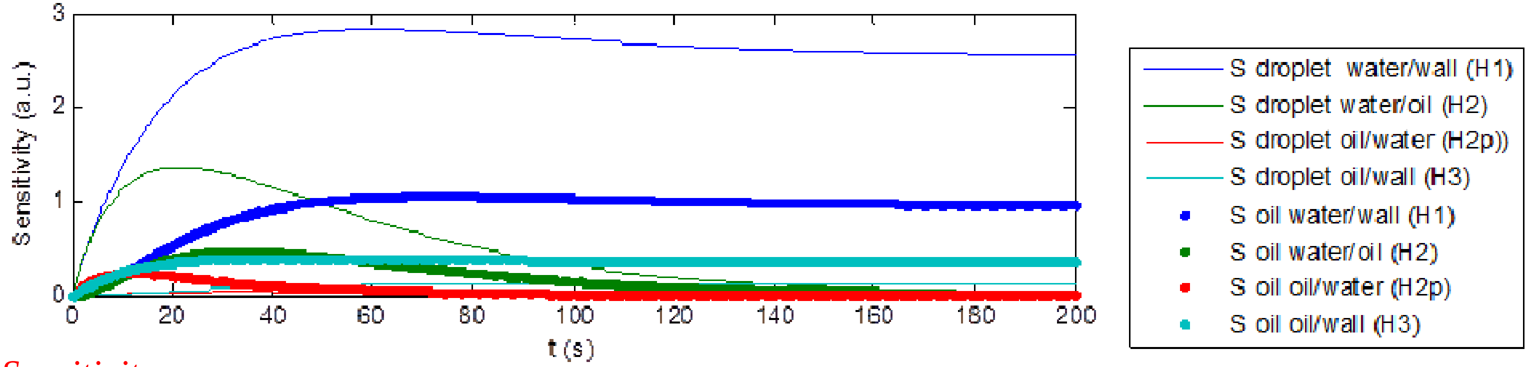
Sensitivity study $S_\beta = \frac{\partial T}{\partial \beta} \times \frac{1}{\beta}$

Water at T0=50°C, oil and plate at Tp=25°C

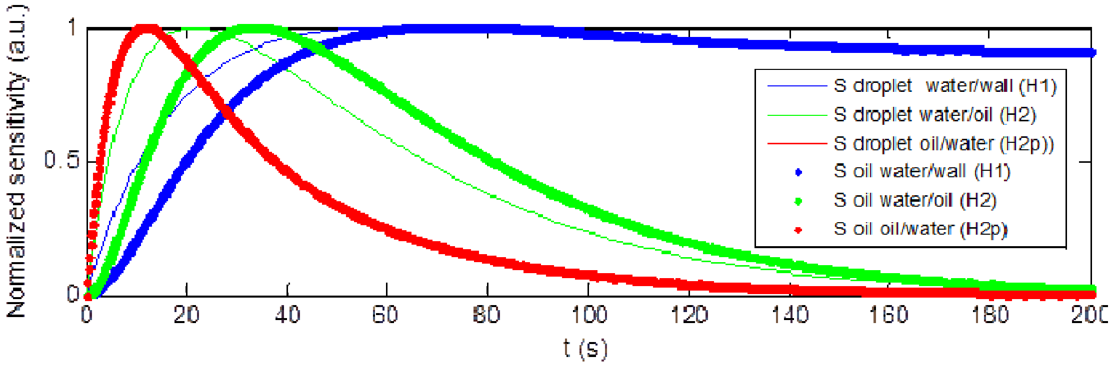
Calculated temperatures



Sensitivity



Normalized Sensitivity



Good sensitivity to heat exchange between oil and water

Validation of the inverse method on noisy analytical solution

Water at T0, oil and plate at Tp

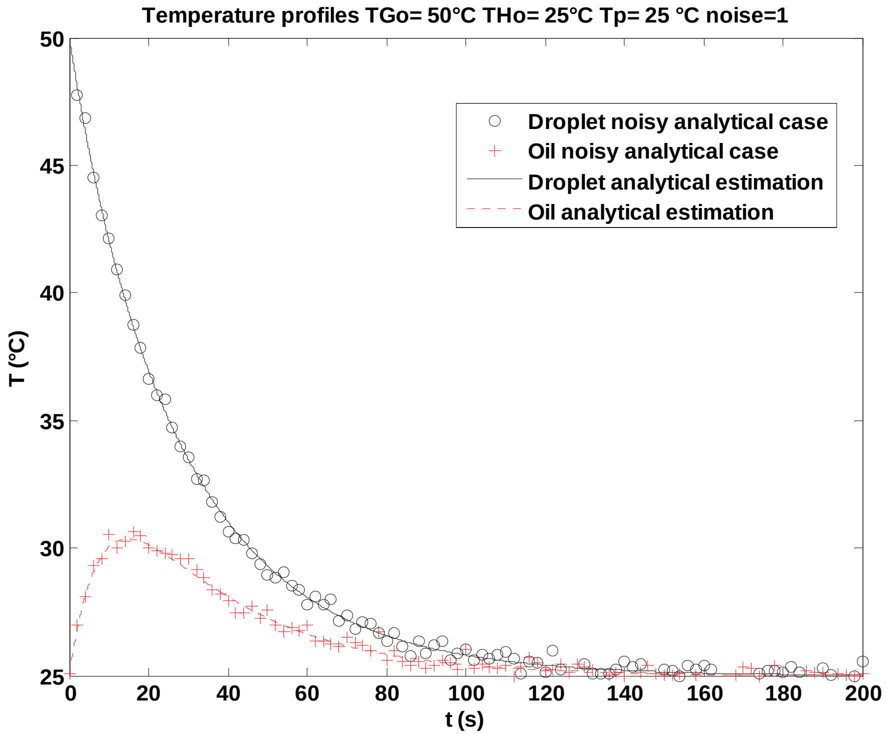
Inverse problem formulation

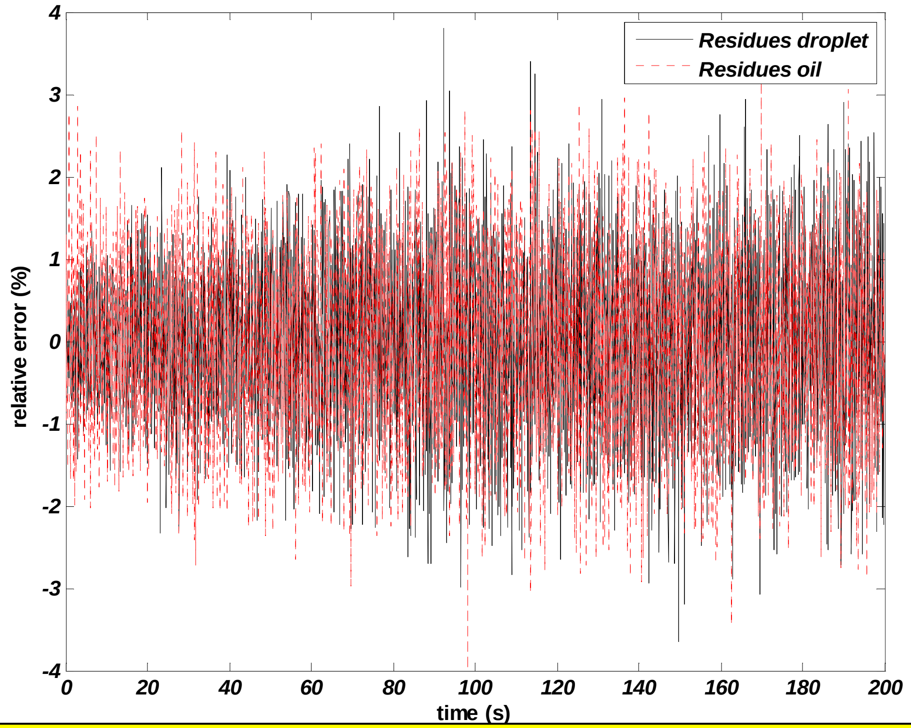
$$X_H = \int_0^t T_H\,dt \quad ; \quad X_G = \int_0^t T_G\,dt$$

$$T_G = \left[1 \quad X_G \quad X_H \quad t\right]\left[T_{G0} \quad (H_1 + H_2) \quad -H_2 \quad -H_1 T_p\right]^T$$

Gauss Markov inversion

$$P = \left({}^T[X][X]\right)^{-1} \cdot {}^T[X]Y$$

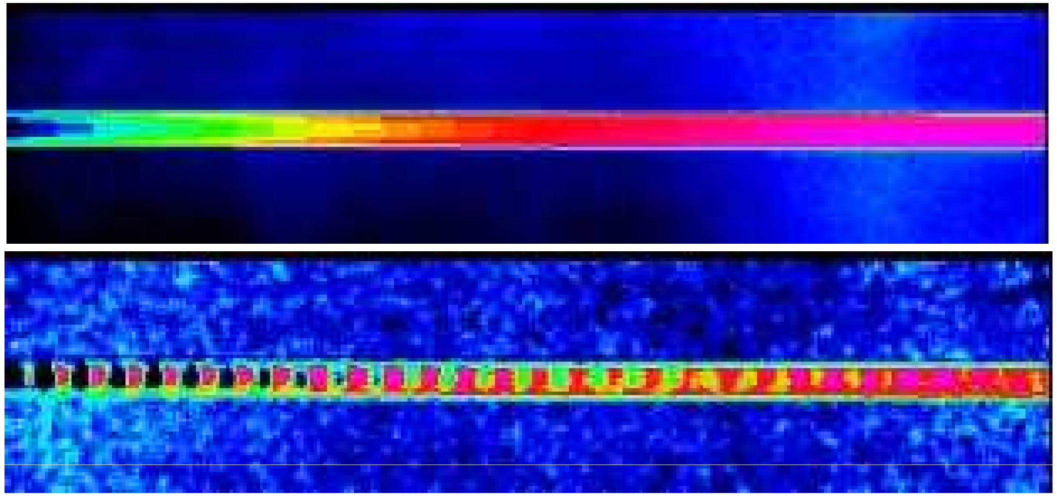




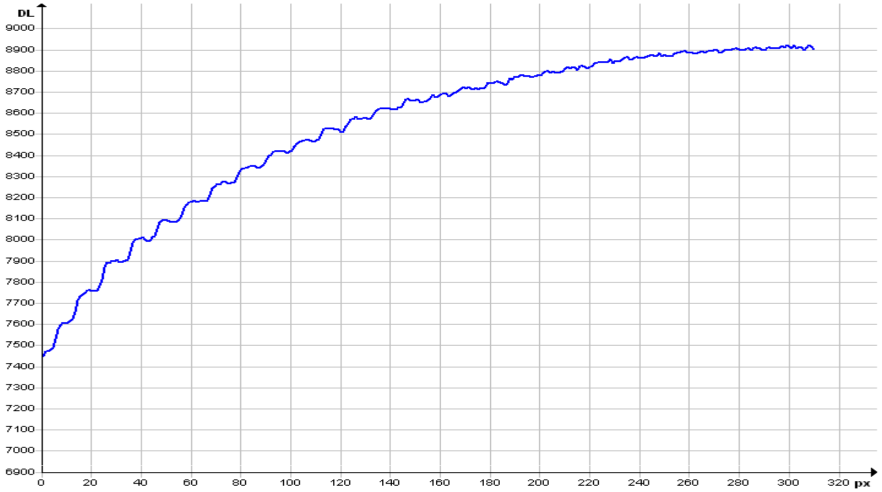
Error lower than 1% on all the parameters

Fluids at the same temperature different from the plate

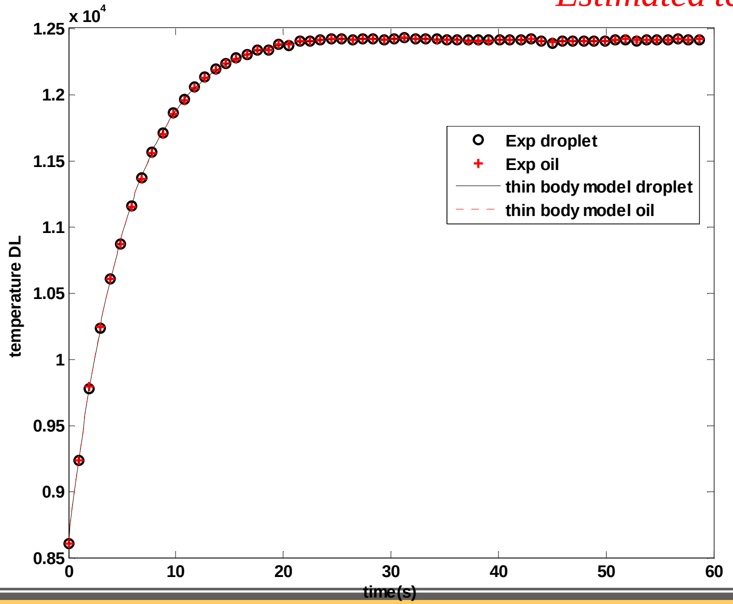
Measured temperature

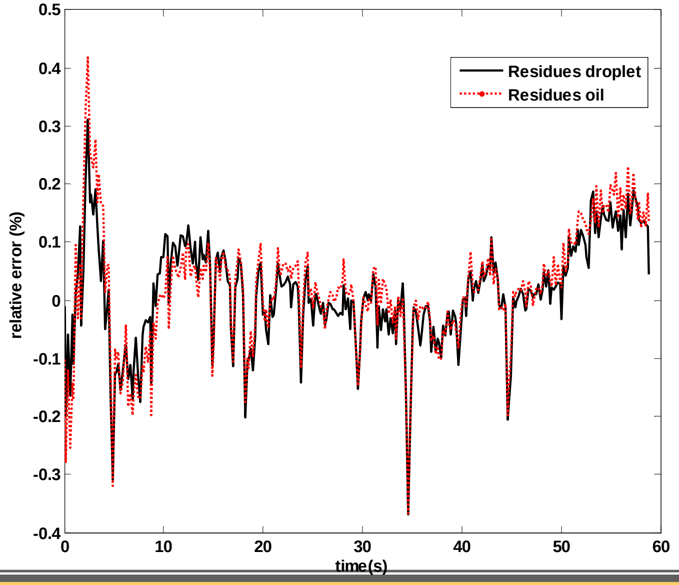


Profil along the channel at ti



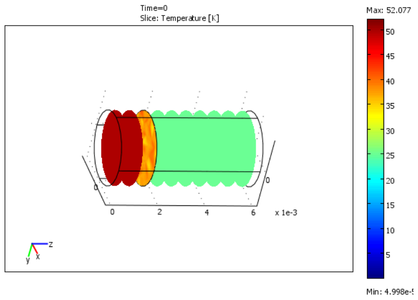
Estimated temperature and residu





Good estimation of the parameters

1Dt homogenised thermal model

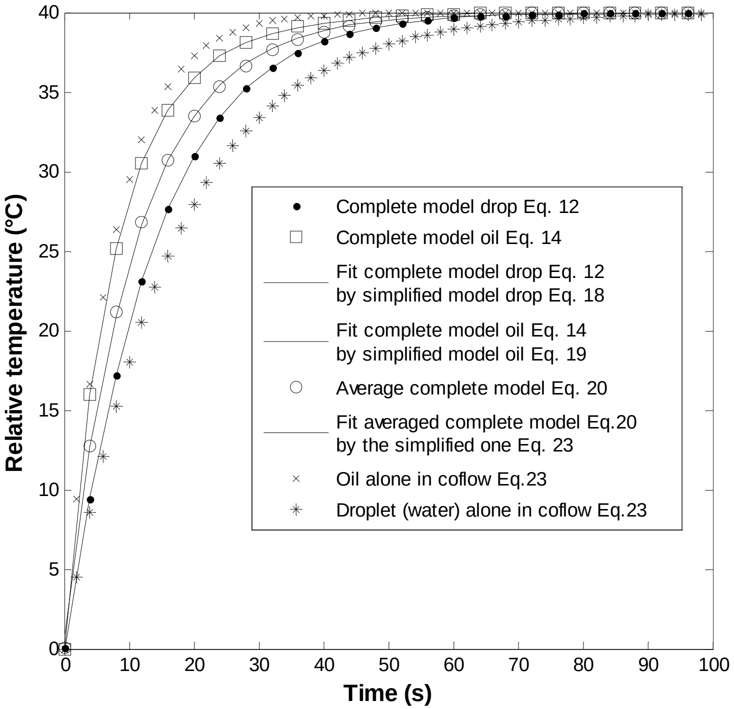


Averaging of the 2Temperature models

$$\frac{d\overline{T}}{dz} = -H(\overline{T}_z - T_p)$$

$$H = \frac{h_p \pi d L_T}{\rho C_p{}^* V U}; \rho C_p{}^* V *= \rho_G C_{pG} V_G + \rho_H C_{pH} V_H \quad ; \quad U = Q_T/S$$

Resolution in Laplace domain



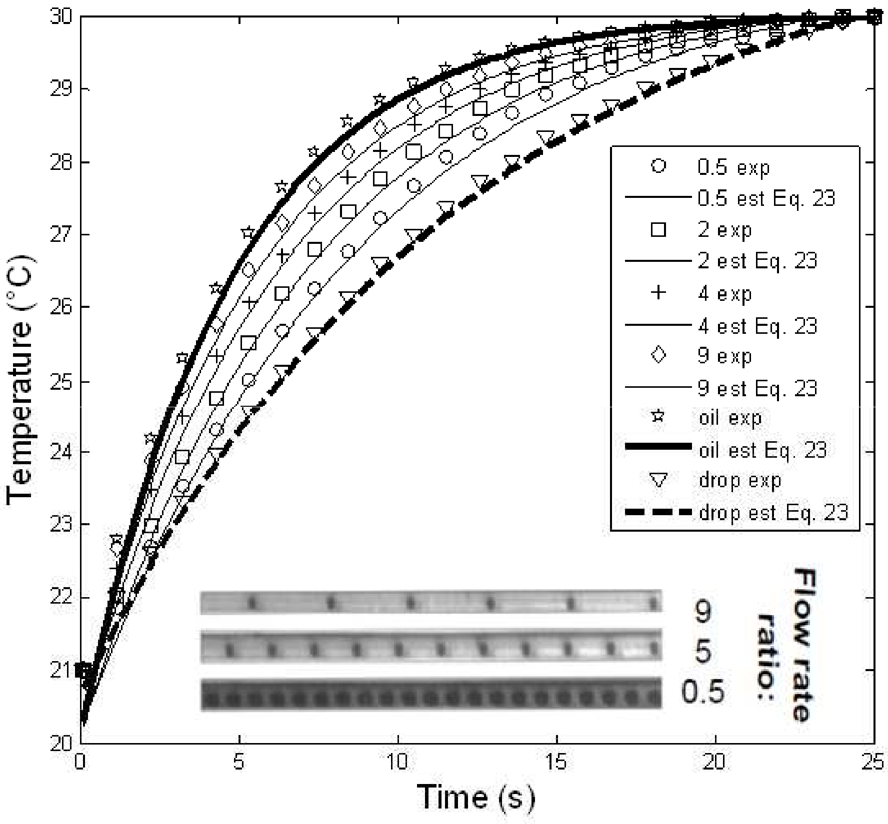
Two limits (oil and water)
2T, two kind of heat exchange
(wall + interface)
1DT, Mixing law

Very simple method for: Estimation of the heat exchange

Influence of the flow rate ratio on heat exchange

Various total flow rate for one oil and water ratio

Same total flow rate with several ratio between oil and water



The mean velocity varies linearly with the total flow rate
This velocity is independent of the oil and water flow rate ratio

Behavior law for the H coefficients

Relationship

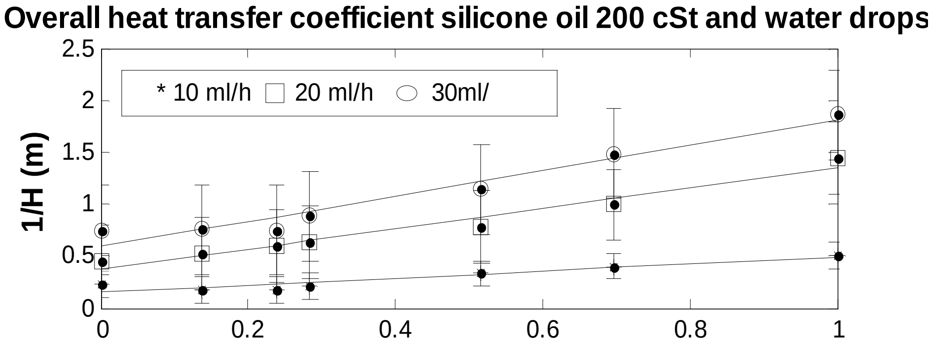
$$\frac{1}{H} = Su + O \text{ with: } S = \frac{1-K}{K_0} \text{ and } O = \frac{K}{K_0}$$

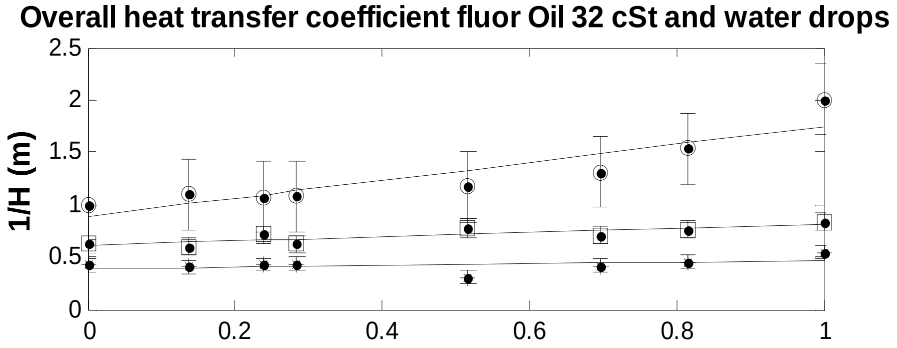
$$K_0 = \frac{\pi d h_p}{Q_T(\rho C_p)_G} \; ; \; K = \frac{(\rho C_p)_G}{(\rho C_p)_H} \; ; \; \alpha = \frac{L_H}{L_G} ; \; u = \frac{1}{1+\alpha}$$

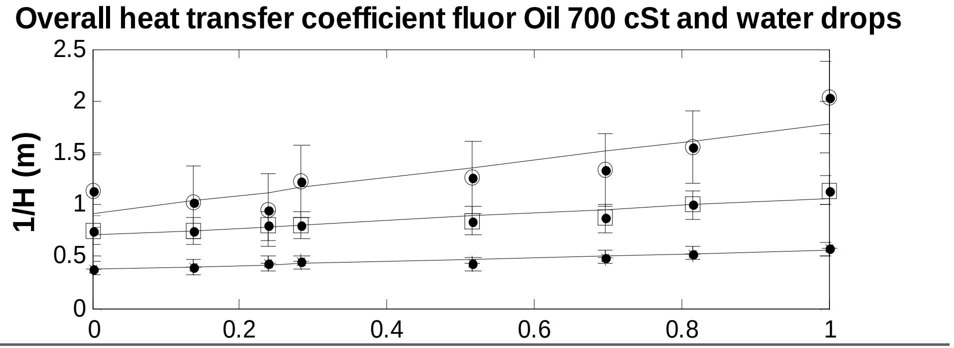
Thermal properties estimation

Oil	Data from supplier		Estimation		Absolute error
	K	Oil ρC_p ($J.m^{-3}.K^{-1}$)	K	Oil ρC_p ($J.m^{-3}.K^{-1}$)	
Silicone oil 200 cSt	3.31	1.2628 $x10^6$	3.20	1.3062 $x10^6$	3%
Fluorinated oil 32 cSt	2.23	1.8744 $x10^6$	2.14	1.9533 $x10^6$	4%
Fluorinated oil 700 cSt	2.11	1.9810 $x10^6$	2.13	1.9624 $x10^6$	1%

Estimated H as function of ratio and fluids


Overall heat transfer coefficient silicone oil 200 cSt and water drops


Overall heat transfer coefficient fluor Oil 32 cSt and water drops


Overall heat transfer coefficient fluor Oil 700 cSt and water drops

First results show that a simplified 2T thin body model is a good representation of droplets thermal behaviour in millifluidic reactor.

Nevertheless in isoperibolic conditions, a 1Dt homogenized model is enough

Advantages

A simple calibration of the fluid heat transfer is necessary for the study of chemical reaction

Correlation could be obtained versus flow rate ratio

Improvements

Make the calibration with different inlet temperature for oil and water (see sensitivity study)

Develop heterodyne technique to enhance the measurement

Validate the obtained heat exchange coefficient versus classical correlations

Performed slow kinetic chemical reaction