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Mathematical Investigation and CFD Simulation of Monolith Reactors: Catalytic Combustion of Methane

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Why micro-fabrication?

- The high heat and mass transfer rates possible in micro-fluidic systems allow reactions to be performed under more aggressive conditions with higher yields than can be achieved with conventional reactors
- New reaction pathways deemed too difficult in conventional microscopic equipment, e.g., direct fluorination of aromatic compounds (Chambers & Spink, 1999), could be pursued.
- Scale-up to production by replication of micro-reactor units used in the lab eliminates costly redesign and pilot plant experiments, thereby shortening the development time

Why micro-fabrication? (contd.)

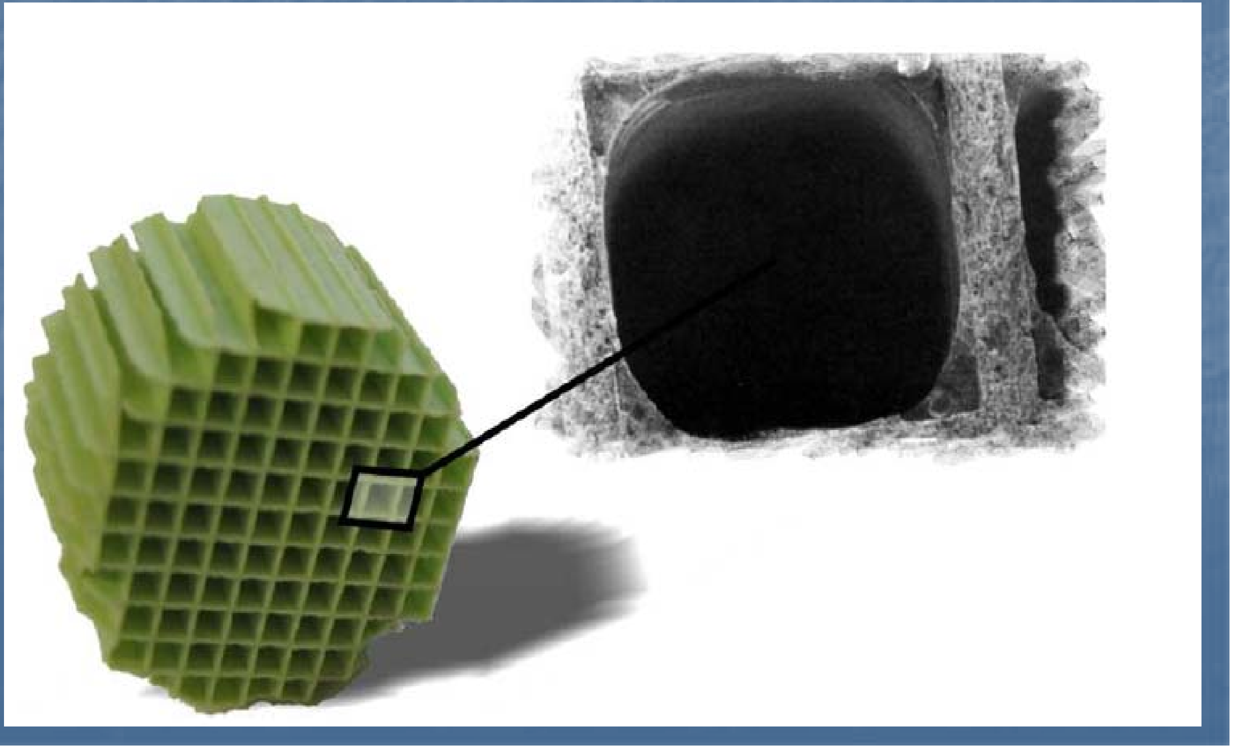
- The presence of integrated sensor and control units could allow the failed reactor to be isolated and replaced while other parallel units continued production.
- These systems are capable of integrating all stages of a complete analysis, including sampling, sample pretreatment, chemical reaction, separation, detection, and data processing in a highly automated and efficient manner.

Ref: Jakeway, S. C.; de Mello, A. J.; Russell, E. L., Miniaturized total analysis systems for biological analysis. *Fresenius' Journal of Analytical Chemistry* **2000,** 366, (6-7), 525-539.

What is this problem?

- A two-phase (gas & solid) transient catalytic combustor model using a simplified flow field inside a single channel to test the advantages of COMSOL Multiphysics software.

Ref: R. E. Hayes and S. T. Kolaczkowski, *Introduction to Catalytic Combustion*. Amsterdam: Gordon and Breach Science Publ., **1997**



Catalytic Combustion

- The complete oxidation of a combustible compound on the surface of a catalyst.
- A flameless process occurring at lower temperatures and, therefore, emitting less nitrogen oxides (Hayes et al. 1997)
- Catalysed combustion offers fewer constraints concerning flammability limits and reactor design.
- The design of the catalytic combustion stage typically calls for monolith systems that offer high surface area but low-pressure drop. The monolith honeycombs are often made of cordierite coated with catalytically active material, whereby a washcoat, mostly alumina, is frequently used to enlarge the surface area.

$$CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O$$

$$\Delta H_{reac} = 802368 + 0.0133T^2 - 14.625T$$

Why COMSOL?

- It has an integrated modeling environment.
- It takes a semi-analytic approach: You specify equations, COMSOL symbolically assembles FEM matrices and organizes the bookkeeping.
- COMSOL is built on top of MATLAB, so user defined programming for the modeling, organizing the computation, or the post-processing has full functionality.
- It provides pre-built templates as Application Modes
- It provides multi-physics modeling linking well known “application modes” transparently.
- COMSOL innovated extended multi-physics-coupling between logically distinct domains and models that permits simultaneous solution.

Assumptions

- The channel is cylindrical and the flow is axisymmetric and Laminar
- The porous medium is homogeneous and isotropic
- The interactions between the porous medium and the clear fluid is simulated by the Brinkman formulation [13]
- The solid matrix and the fluid are assumed to be at local thermal and concentration equilibrium with each other
- Homogeneous reaction and heat radiation in the bulk phase are ignored.

Mathematical Presentation

$$\frac{\partial \rho u}{\partial z}+\frac{1}{r}\frac{\partial(\rho v)}{\partial r}=0 \qquad (1$$

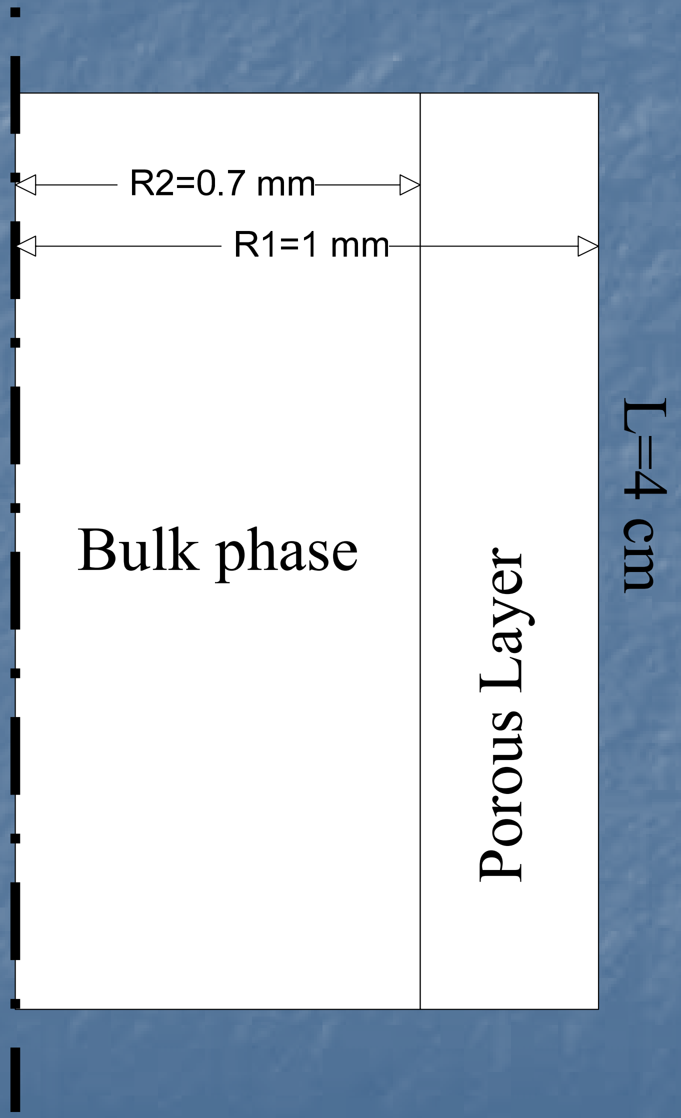
$$\rho u\frac{\partial u}{\partial z}+\rho v\frac{\partial(u)}{\partial r}=-\frac{\partial p}{\partial z}+\frac{\partial}{\partial z}\left[2\mu\frac{\partial u}{\partial z}-\frac{2}{3}\mu\nabla .V\right] \qquad (2$$

$$+\frac{1}{r}\frac{\partial}{\partial r}\left[\mu r\left(\frac{\partial v}{\partial z}+\frac{\partial u}{\partial r}\right)\right]$$

$$\rho u\frac{\partial v}{\partial z}+\rho v\frac{\partial(v)}{\partial r}=-\frac{\partial p}{\partial z}+\frac{\partial}{\partial z}\left[\mu\left(\frac{\partial v}{\partial z}+\frac{\partial u}{\partial r}\right)\right] \qquad (3$$

$$+\frac{\partial}{\partial z}\left[2\mu\frac{\partial v}{\partial r}-\frac{2}{3}\mu\nabla .V\right]$$

$$+\frac{2\mu}{r}\frac{\partial}{\partial r}\left[\frac{\partial v}{\partial z}-\frac{v}{r}\right]$$



Mathematical Presentation (contd.)

$$\rho u \frac{\partial Y_k}{\partial z} + \rho v \frac{\partial Y_k}{\partial r} = \left(\frac{\partial J_{k,z}}{\partial z} + \frac{1}{r} \frac{\partial (r J_{k,z})}{\partial r} \right) + \dot{\omega}_k W_k \qquad (4$$

$$(k = 1, \ldots, K_g)$$

$$\begin{aligned} \rho c_p \left(u \frac{\partial T}{\partial z} + v \frac{\partial T}{\partial r} \right) &= \left(u \frac{\partial p}{\partial z} + v \frac{\partial p}{\partial r} \right) \qquad (5 \\ &+ \frac{\partial}{\partial z} \left(\lambda \frac{\partial T}{\partial z} \right) + \frac{\partial}{\partial r} \left(r \lambda \frac{\partial T}{\partial r} \right) \\ &- \sum_{k=1}^{K} c_{pk} \left(J_{kz} \frac{\partial T}{\partial z} + J_{kr} \frac{\partial T}{\partial r} \right) \\ &- \sum_{k=1}^{K} h_k \dot{\omega}_k W_k \end{aligned}$$

Boundary conditions

- **At the inlet of the channel:**
  Initial values for Velocity, Temperature and Concentration

- **At the axisymmetric line of the channel:**
  Axial symmetry for all parameters

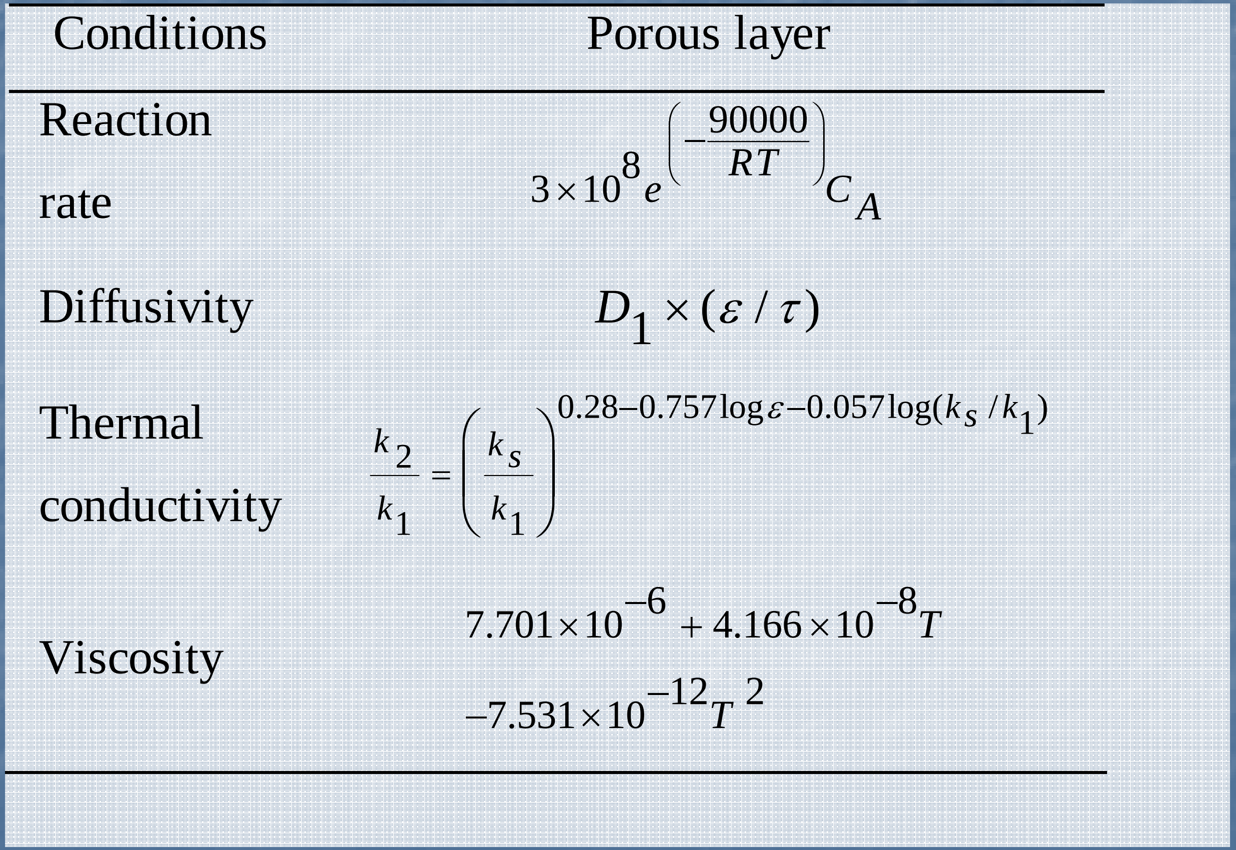
- **At the outlet of the channel:**
  Convective flux is assumed

- **At the wall:**
  No slip condition is assumed

Simulation parameters for the bulk phase

Conditions	Bulk Phase
Reaction rate	0
Diffusivity	$D_1 = 9.99 \times 10^{-5} \times (T^{1.75} / P)$
Thermal conductivity	$k = 1.679 \times 10^{-2} + 5.073 \times 10^{-5} T$
Viscosity	$7.701 \times 10^{-6} + 4.166 \times 10^{-8} T$ $-7.531 \times 10^{-12} T^2$

Simulation parameters for the porous layer

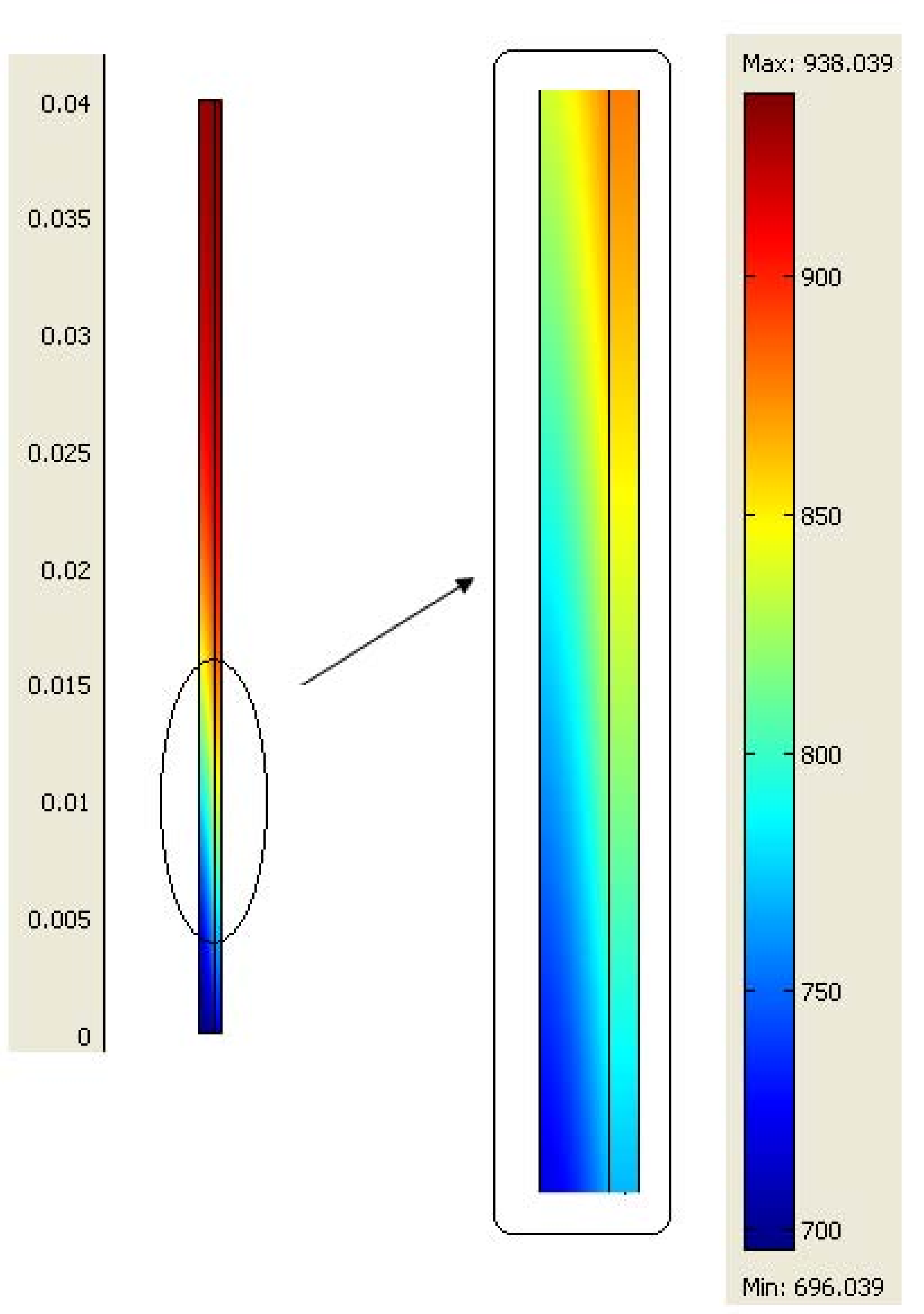


Conditions	Porous layer
Reaction rate	$3\times10^{8}e^{\left(-\frac{90000}{RT}\right)}C_A$
Diffusivity	$D_1\times(\varepsilon/\tau)$
Thermal conductivity	$\frac{k_2}{k_1}=\left(\frac{k_s}{k_1}\right)^{0.28-0.757\log\varepsilon-0.057\log(k_s/k_1)}$
Viscosity	$7.701\times10^{-6}+4.166\times10^{-8}T$ $-7.531\times10^{-12}T^{2}$

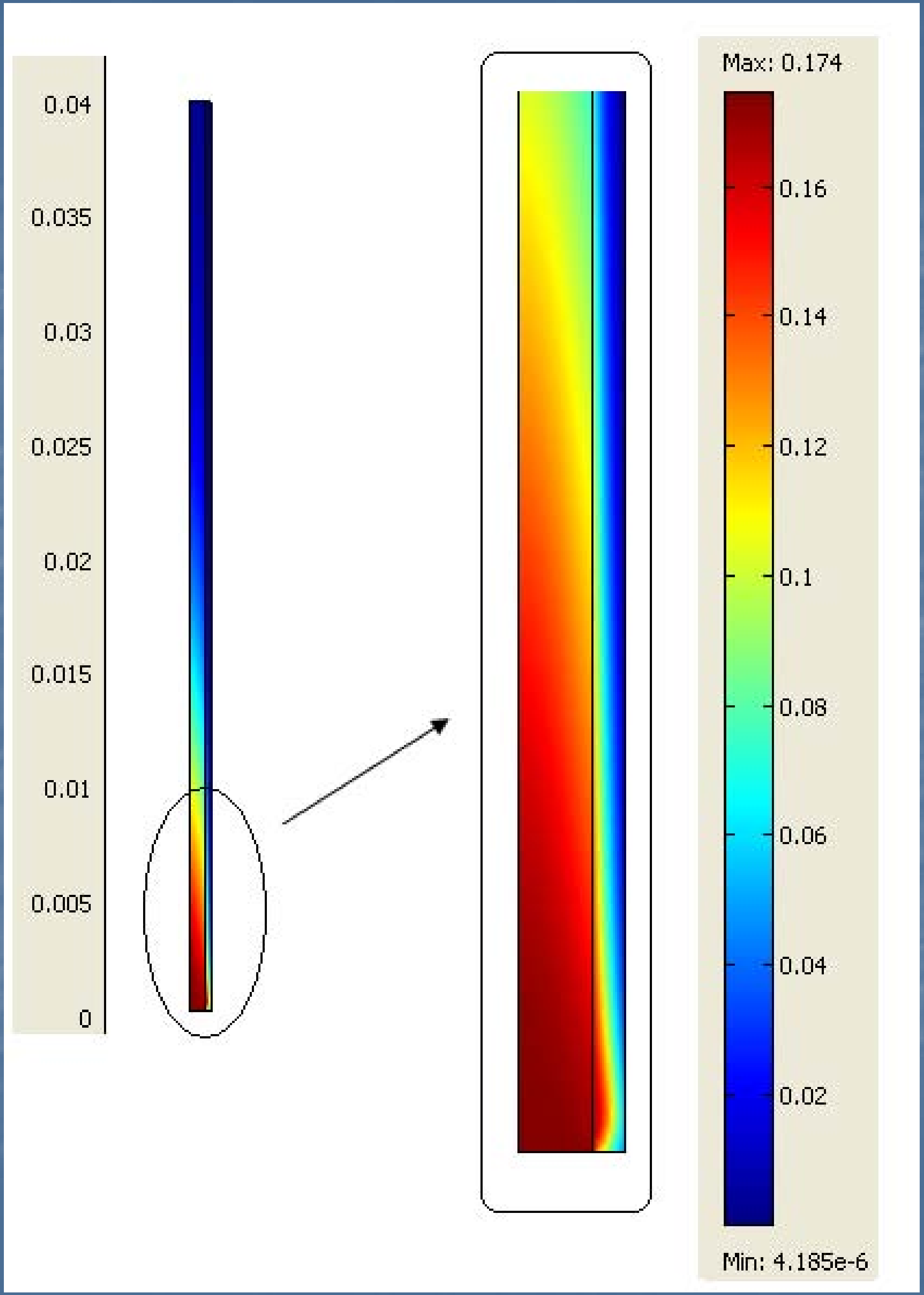
Simulation conditions

Geometrical Conditions	
Channel length (m)	0.04
Porous layer thickness (mm)	1.0
Catalyst support materials	
Tortuosity, τ	4
Porosity, ε	0.4
Permeability, K m^2	1×10^{-8}
Thermal conductivity, k_s W/mK	25
Heat capacity, C_{ps} $J/kg.K$	900
Density, ρ_s kg/m^3	7870

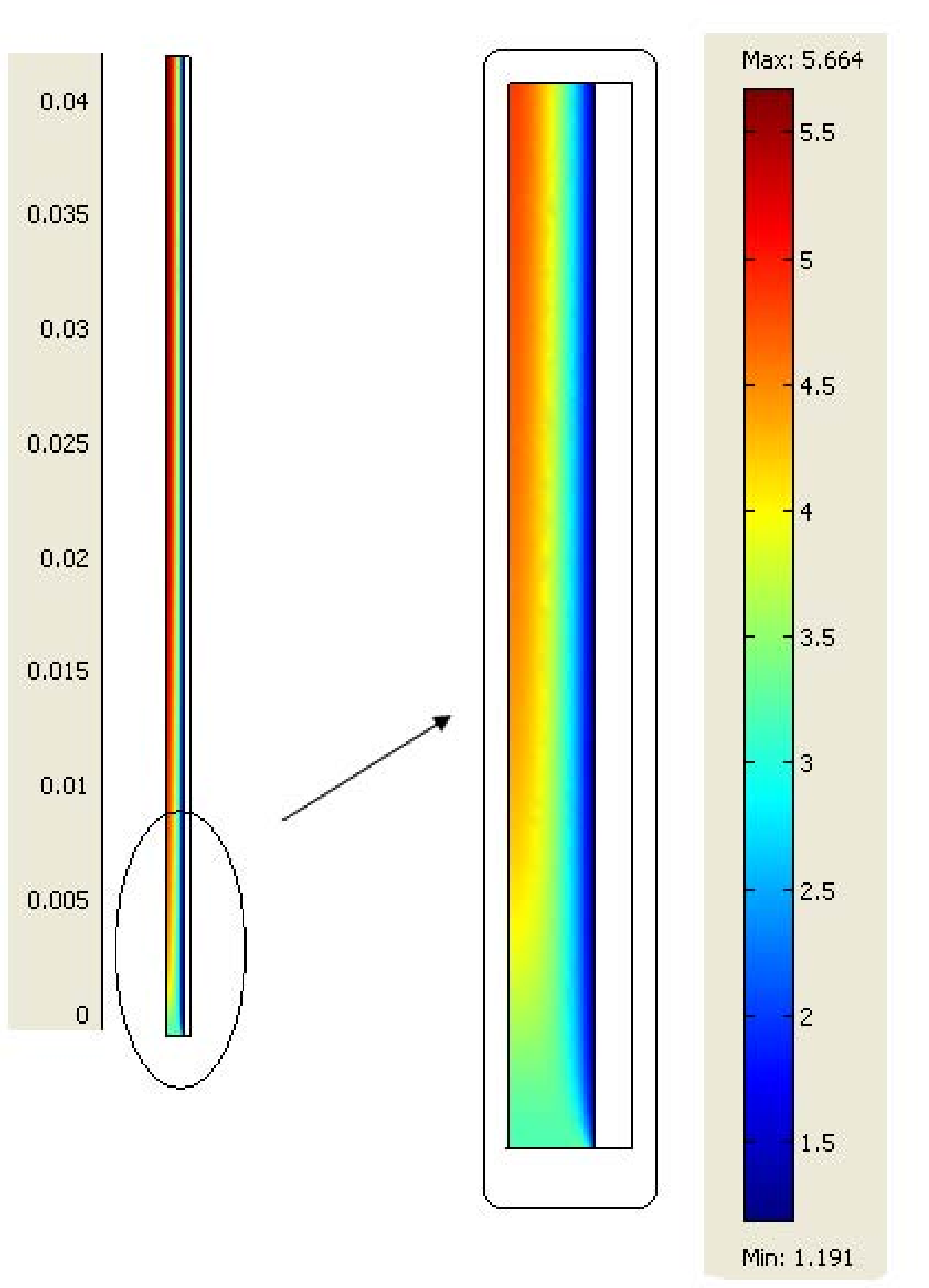
Temperature profile along the channel



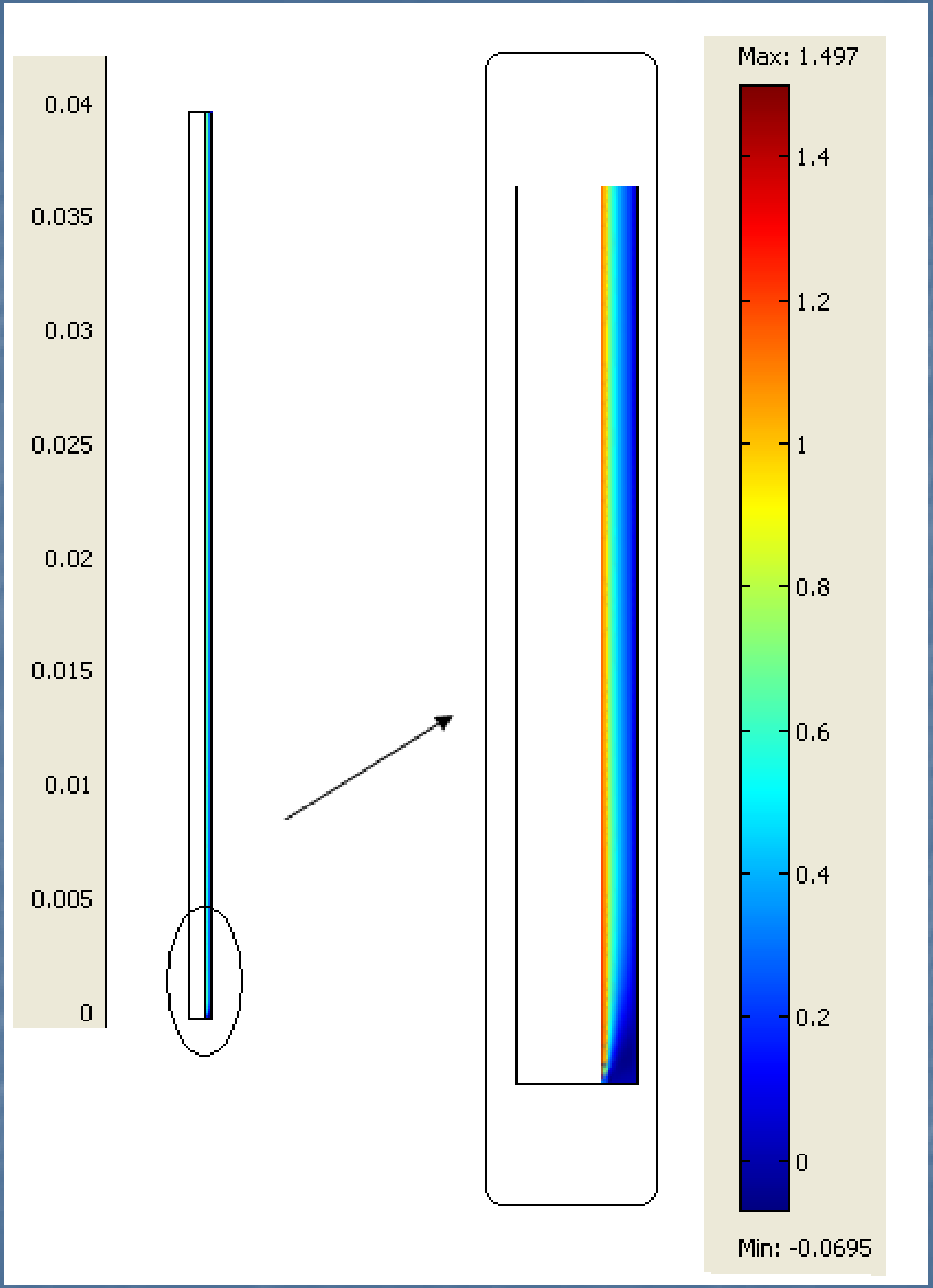
Concentration profile along the channel



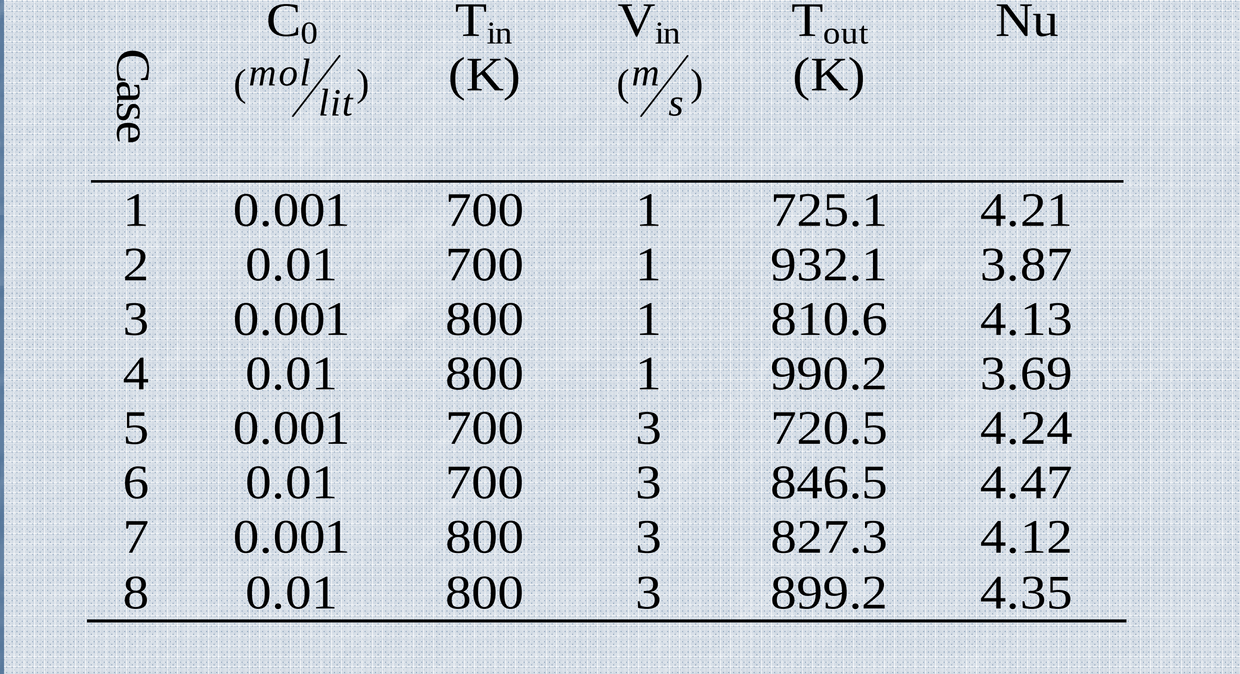
Velocity profile in the bulk phase



Velocity profile in the porous layer



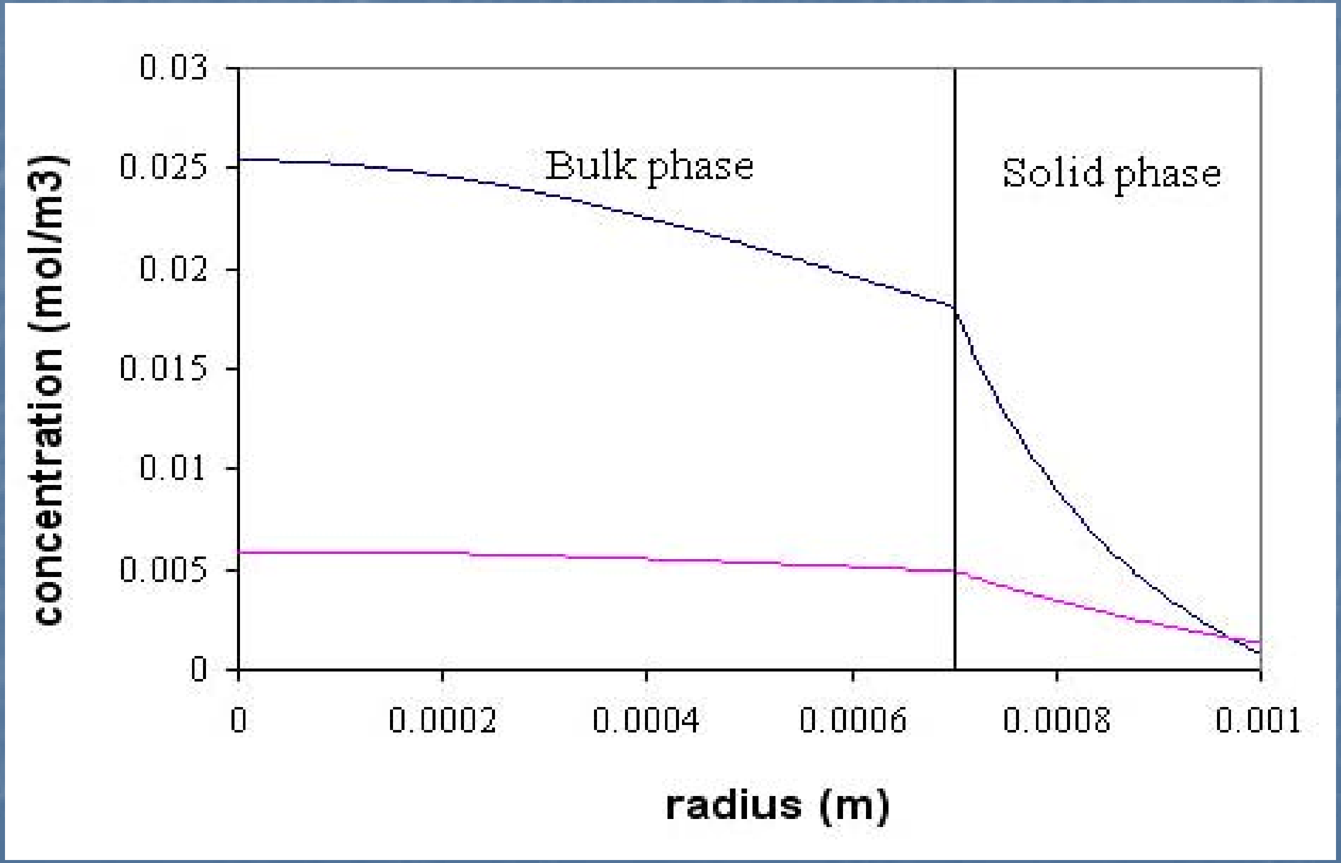
Summary of conditions in the simulations



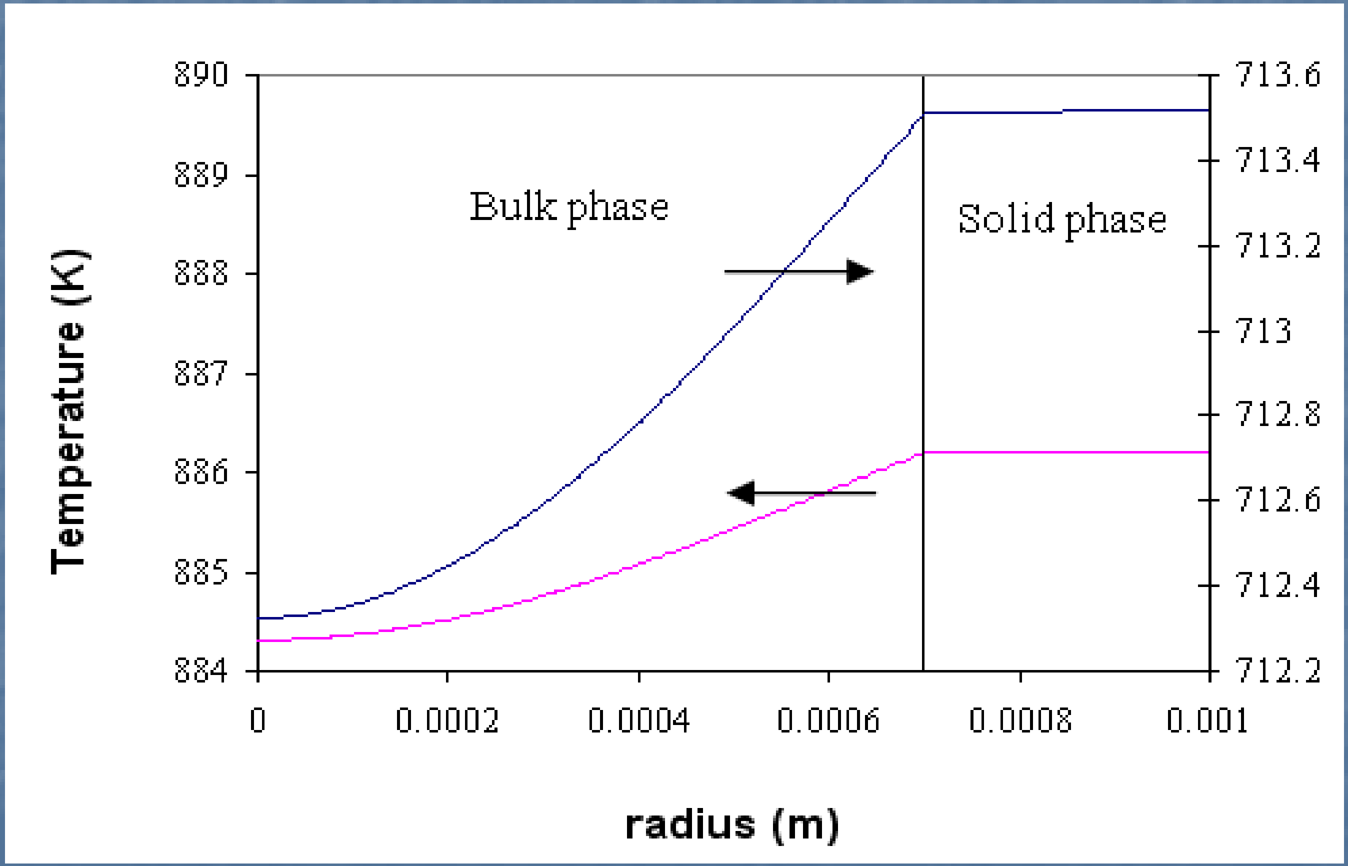
Case	C_0 $(\mathrm{mol}/\mathrm{lit})$	T_{in} (K)	V_{in} (m/s)	T_{out} (K)	Nu
1	0.001	700	1	725.1	4.21
2	0.01	700	1	932.1	3.87
3	0.001	800	1	810.6	4.13
4	0.01	800	1	990.2	3.69
5	0.001	700	3	720.5	4.24
6	0.01	700	3	846.5	4.47
7	0.001	800	3	827.3	4.12
8	0.01	800	3	899.2	4.35

$$Nu = \frac{Convective\ heat\ transfer}{Conductive\ heat\ transfer}$$

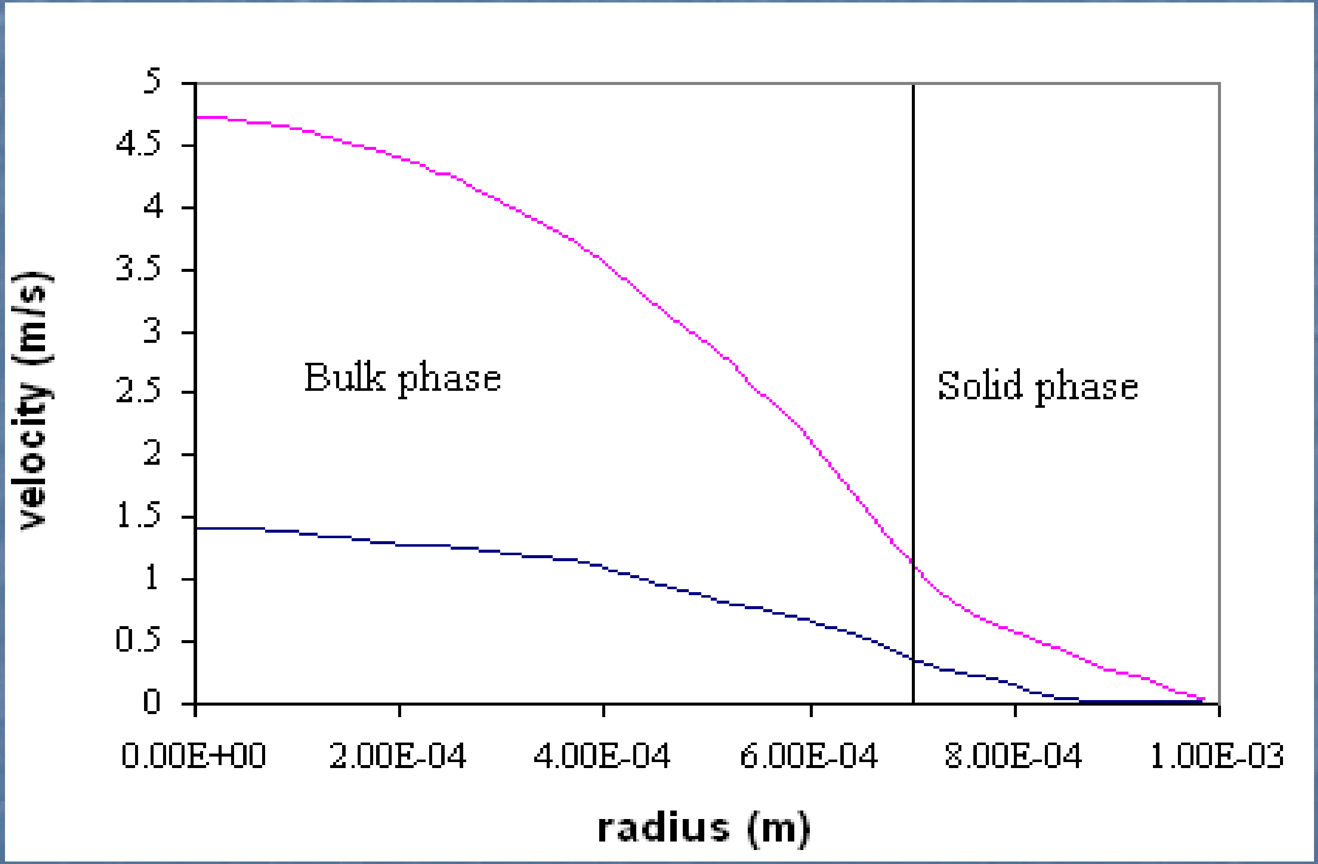
Concentration profile for cases 1 (up) and 2 (down)



Temperature profile for cases 1 (up) and 4 (down)



Velocity profile for cases 1 (up) and 4 (down)



Thanks for your attention