DECHEMA-Kolloquium
Organische Synthesen in Mikrostrukturreaktoren
Grundlagen des Wärme- und Stofftransports in Mikrostrukturreaktoren
-
Fundamentals of Heat and Mass Transfer in Microreactors

8th November 2007

Dr.-Ing. Norbert Kockmann

Design of Microsystems,
Department of Microsystems Engineering – IMTEK,
Albert-Ludwig University of Freiburg

now at: Lonza AG, CH-3930 Visp, R&D Exclusive Synthesis

Intro
Fluid properties
Fluid dynamics
Heat transfer
Mass transfer
Chemical reactions
Conclusions

Contents

Fundamentals of Heat and Mass Transfer

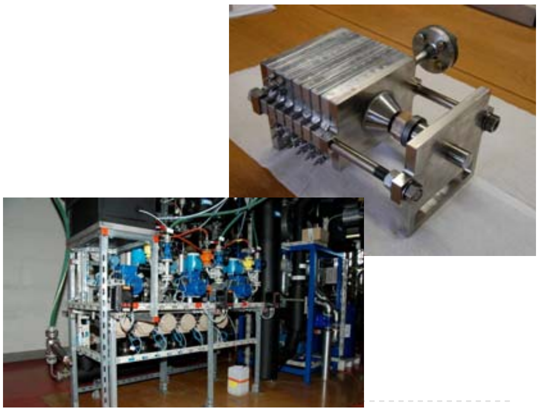
- Introduction and motivation
- Scaling and fluid properties
- Fluid dynamics
- Pressure loss
- Heat transfer in straight channels
- Heat transfer in curved channels and networks
- Convective mixing in microchannels
- Chemical reactions and transport processes
- Conclusions
- References

University of Freiburg, IMTEK

- IMTEK: 18 professor ships
- approx. 300 employees
- approx. 500 students, diploma, bachelor, master
- 9300 m^2 laboratories and bureaus
- 600 m^2 clean room

Microreactors at Lonza, Visp

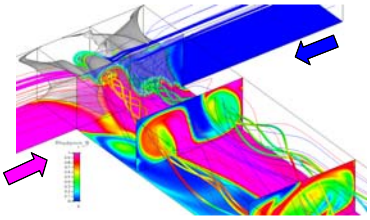
- Area 90 ha
- Employees ~2700
- Plants
  - Naphtha cracker
  - Single product, multi-product and multipurpose facilities
  - Fully integrated waste management
- Products
  - Active pharmaceutical ingredients and Biopharmaceuticals)
  - Vitamines (Vitamin B_3)
  - Peptides and oligonucleotides
  - …
- Microreactors
  - pioneering work of D. Roberge
  - 3 modules for various reactions
  - GMP production



Microfluidics and Micro Process Engineering

Main characteristics

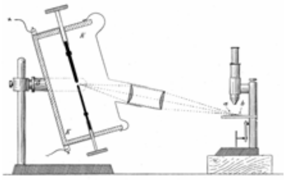
- small channels: d_h from ca. 10 µm to 1 000 µm
- cross sections: rectangular, trapezoidal, semi-circular
- often laminar flow: $\mathrm{Re} = \frac{w\,d_h}{\nu} < 1000$



- Analysis of small volumes ⇒ **µTAS** (micro total analysis systems)
- Process design in microstructures ⇒ **Lab-on-a-chip**
- Production of chemicals in small amounts ⇒ **µVT, MPE**

EMICH
LEHRBUCH
DER MIKROCHEMIE

WIESBADEN.
VERLAG VON J. F. BERGMANN.
1911.

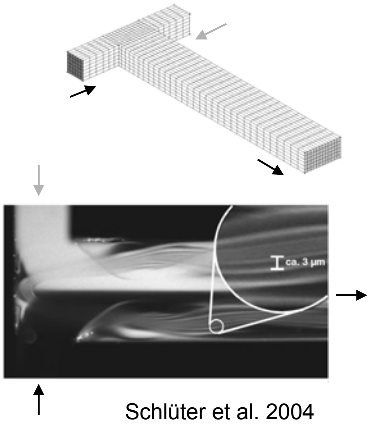


© MicroChemTec backbone

Microfluidics and Micro Process Engineering

Why micro process engineering?

- Defined flow regimes and properties
  - mainly laminar flow
  - defined residence time and distribution
  - small hold-up
- Augmented transport processes
  - rapid mixing
  - high heat transfer
  - flame suppression
  - small inertia and capacities
  - integrated processes and sensors

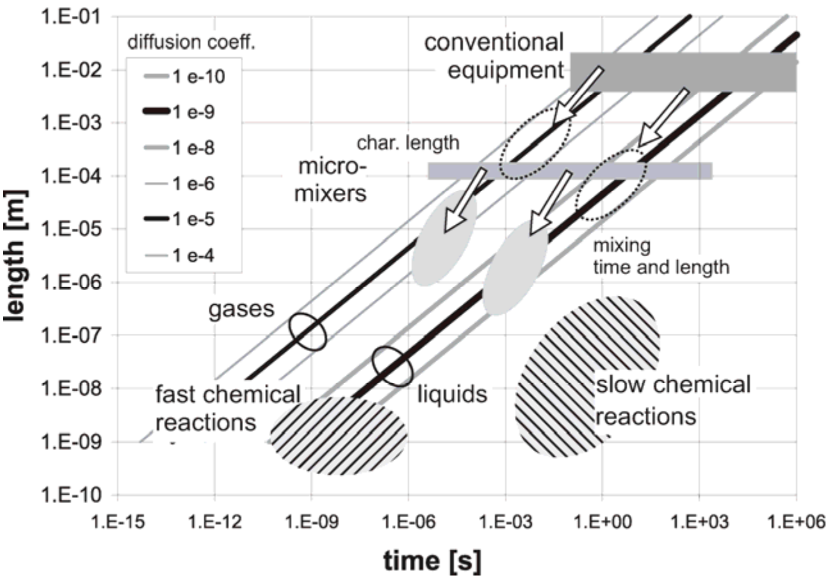


Schlüter et al. 2004

- Frequently questions to be answered
  - Why has this device these dimensions and materials?
  - Why do I operate under these conditions?
    (temperature, pressure, concentrations, …)
  - Why do I need these solvents?
  - Why do I need this reaction time and constellation?

Length scales

- typical regimes in conventional and microstructured devices

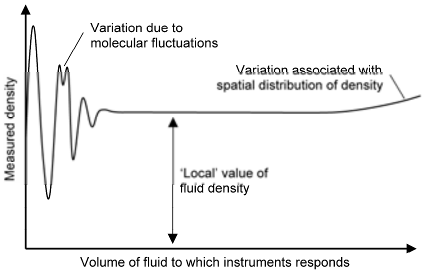


Kockmann 2007

- diffusion in liquids is slow
- fast convective mixing

Fluid behavior

- Fluid properties and control volume

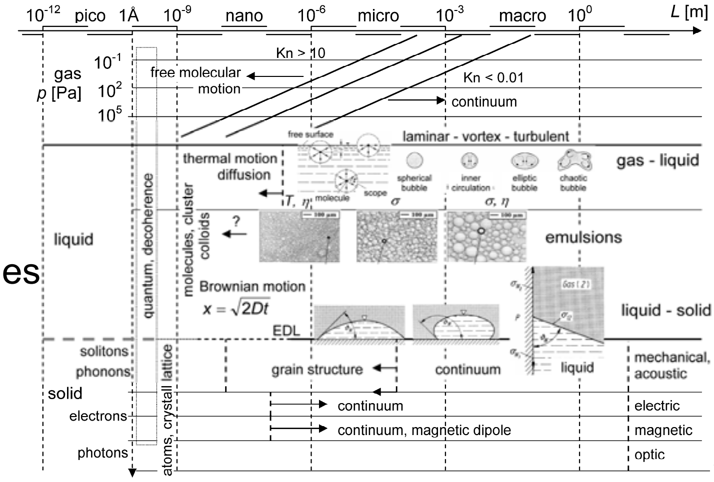


Batchelor, 2000

- large thermal fluctuations for only few molecules,
- classical properties at $N > 10^6$,
- fluctuations are also possible in large systems.

- Phase limits
  - number of gas molecules
  - free surfaces
  - system boundaries

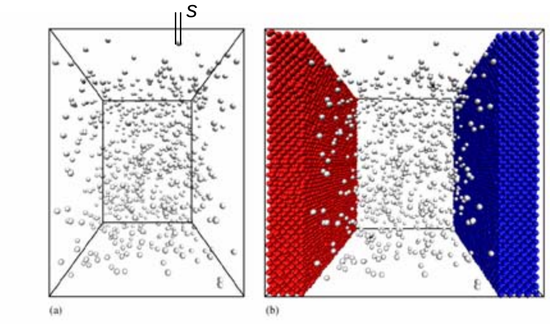
Kockmann 2007



Properties of gases

- Free mean path

$$\Lambda = \frac{\overline{w}\,k\,T}{w_{rel}\,\pi\,s^2\,p} = \frac{k\,T}{\sqrt{2}\,\pi\,s^2\,p}$$



S.Nedea et al. 2006

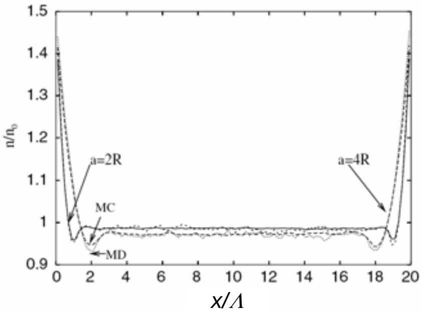
- Knudsen number Kn

$$\mathrm{Kn} = \frac{\Lambda}{L}$$

Knudsen number regimes

	Kn < 0.001	continuum (sometimes 0.01 as limit)
0.001	< Kn < 0.1	rarefied gases
0.1	< Kn < 10	transition regime
10	< Kn	free molecular regime

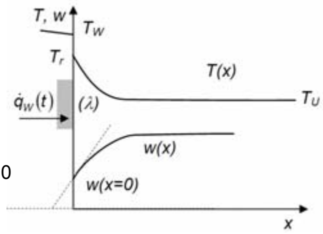
- Molecule behavior at the wall



- Slip length

$$w(x=0) = \zeta \left(\frac{\partial w}{\partial x} \right)_{x=0}$$

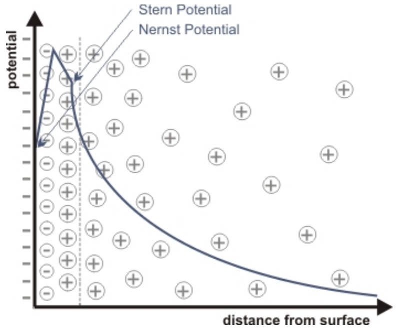
$$\zeta \approx \frac{2-\beta}{\beta}\,\Lambda$$



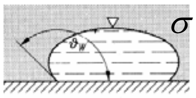
Behavior of liquids

- Number of molecules N > 10^6 gives cube with 100 molecules on corner length: approx. 10 – 30 nm
  → continuum in the range of L > 1 µm

- Surfaces play major role
  Stern layer
  Generation of electrically charged layer
  → electro-kinetic pumps



- hydrophilic / hydrophobic
  Wettability and surface tension
  contact angle



$$L = \sqrt{\frac{\sigma}{g(\rho_l - \rho_v)}}$$ Laplace Länge

$$\mathrm{Co} = \frac{1}{d_h}\sqrt{\frac{4\,\sigma}{g(\rho_l - \rho_v)}}$$ Confinement number

L. Cheng, D. Mewes, 2006

Balance equations

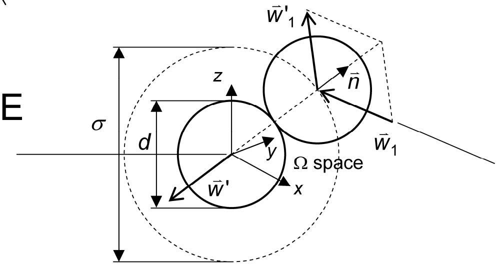
- Overview of balance equations

Fluid dynamics models Gad-el-Hak 2006

molecular
deterministic
Molecular Dynamics
statistical
Liouville
Direct **S**imulation **M**onte **C**arlo
Lattice Boltzmann models

continuum
Euler
Navier-Stokes
Burnett
Chapman - Enskog

- Boltzmann transport equation BTE



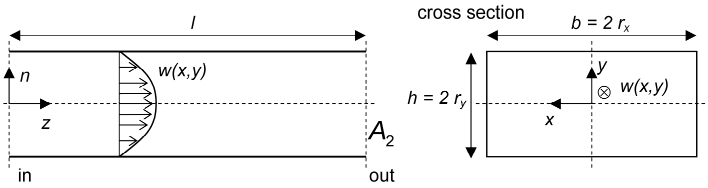
$$\frac{\partial f}{\partial t}+\vec{w}\cdot\frac{\partial f}{\partial \vec{x}}+\frac{1}{m}\quad F\,\frac{\partial f}{\partial \vec{x}}=J_{\text{win}}-J_{\text{loss}}$$

$$\frac{\partial f}{\partial t}+\vec{w}\cdot\frac{\partial f}{\partial \vec{x}}+\frac{1}{m}\quad F\,\frac{\partial f}{\partial \vec{w}}=\frac{\sigma^2}{2}\int\left|\vec{V}\cdot\vec{e}\right|\left(f'\,f_1'-f\,f_1\right)\mathrm{d}\omega\,\mathrm{d}w_{x,1}$$

- balance equations can be derived from BTE
- direct solution only possible for few special cases, Cercignani (1988), Sone (2002)

Conservation Equations

- continuity equation



$$\frac{D\rho}{Dt}+\rho\,\mathrm{div}\,\vec{w}=0$$

- momentum equation
  Navier-Stokes equations

$$\rho\frac{D\vec{w}}{Dt}=\rho\left(\frac{\partial}{\partial t}+\vec{w}\,\mathrm{div}\right)\vec{w}=\rho\,\vec{g}-\mathrm{grad}\,p+\mathrm{Div}\left[\eta\left(2\,\mathrm{grad}\,\mathbf{w}-\frac{2}{3}\vec{\delta}\,\mathrm{div}\,\vec{w}\right)\right]$$

$$\text{with }\mathrm{grad}\,\mathbf{w}=\frac{1}{2}\left[\mathrm{grad}\,\vec{w}+\left(\mathrm{grad}\,\vec{w}\right)^{\mathrm{T}}\right]$$

Reynolds number

$$\mathrm{Re}=\frac{w\,d_h}{\nu}$$

- for long straight channels in *z*-direction

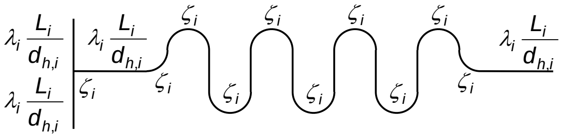
$$\frac{\partial(\rho A)}{\partial t}=0=\frac{\partial(\rho w A)}{\partial z}\qquad \rho w_2 A_2=\rho w_1 A_1$$

$$\frac{\partial(\rho A w)}{\partial t}=-\frac{\partial(\rho w A w)}{\partial z}-\frac{\partial(p A)}{\partial z}+p\frac{\partial A}{\partial z}-\tau L_c-\rho A g$$

- Euler equations for $\eta = 0,\ \tau = 0$

Energy equations

- First law of thermodynamics
  $$\mathrm{d}e = \mathrm{d}e_{kin} + \mathrm{d}e_{pot} + \mathrm{d}u = \mathrm{d}q + \mathrm{d}w_V$$
- energy equation
  $$\frac{\rho}{2} w_2^2 + p_2 - \rho g y_2 = \frac{\rho}{2} w_1^2 + p_1 - \rho g y_1 + w_{t12} - \varphi_{12}$$
  energy dissipation $\varphi_{12} = p_1 - p_2 = \Delta p_{12}$ $\qquad \varepsilon_{12} = \frac{\varphi_{12} \cdot \dot{V}}{m} = \frac{\varphi_{12} \cdot \overline{w}}{\rho \cdot L}$
- kinetic ansatz for pressure loss in channels
  $$\Delta p = \sum \left(\lambda_i \frac{L_i}{d_{h,i}} + \zeta_i \right) \frac{\rho}{2} w_i^2$$



Euler number $\mathrm{Eu} = \frac{\Delta p}{\rho w^2} = \frac{1}{2} \sum \left(\lambda_i \frac{L_i}{d_{h,i}} + \zeta_i \right)$

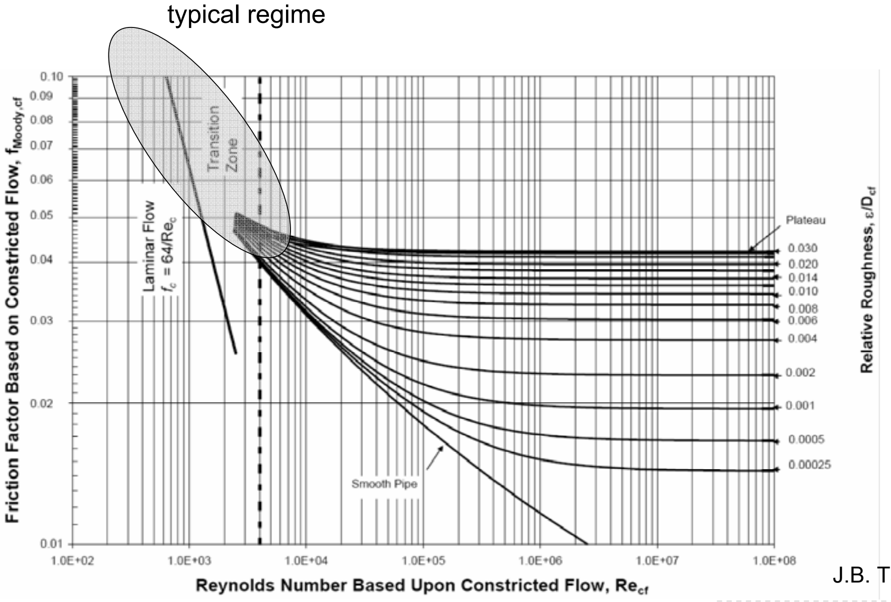
- laminar flow
  $$\lambda_i = \frac{C_{f,i}}{\mathrm{Re}_i} = \frac{C_{f,i} \nu}{w_i \cdot d_{h,i}} \quad \text{and} \quad \Delta p = \sum \left(\frac{\eta C_f L_i}{2 d_{h,i}^2} w_i + \zeta_i \frac{\rho}{2} w_i^2 \right)$$

Pressure loss

- Influence on the flow resistance
  - transition laminar-turbulent
  - available flow cross section

$$\Delta p = \sum \left(\frac{\eta C_f L_i}{2 d_{h,i}^2} w_i + \zeta_i \frac{\rho}{2} w_i^2 \right)$$

- influence on pressure loss → Moody diagram

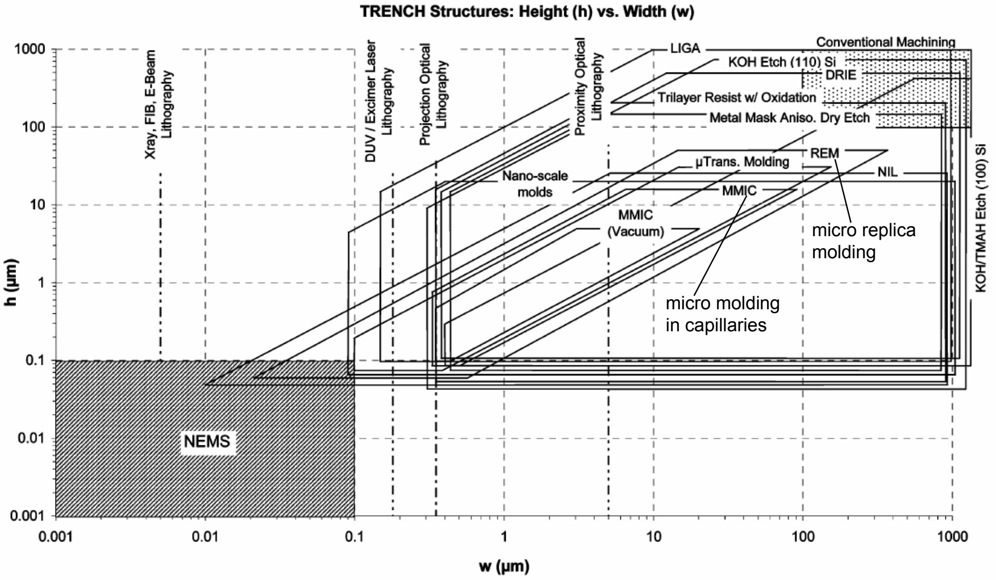


J.B. Taylor et al., 2005

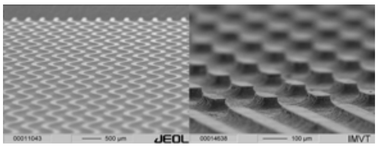
Intro
Fluid properties
Fluid dynamics
Heat transfer
Mass transfer
Chemical reactions
Conclusions

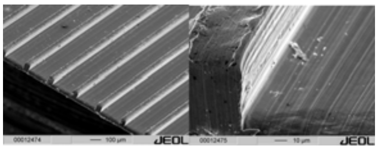
Surface structure and texture

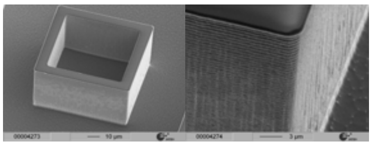
- Fabrication processes, overview from Quinn et al. 2006



Kockmann, 2006,
Chap. 10, 11, 12


isotropic wet-etching of stainless steel

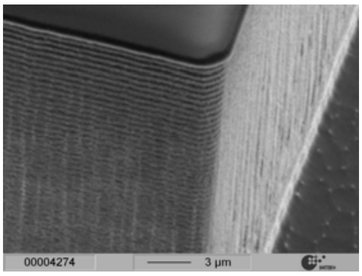

mechanical fabrication of stainless steel


Reactiv ion etching of silicon, DRIE, ASE

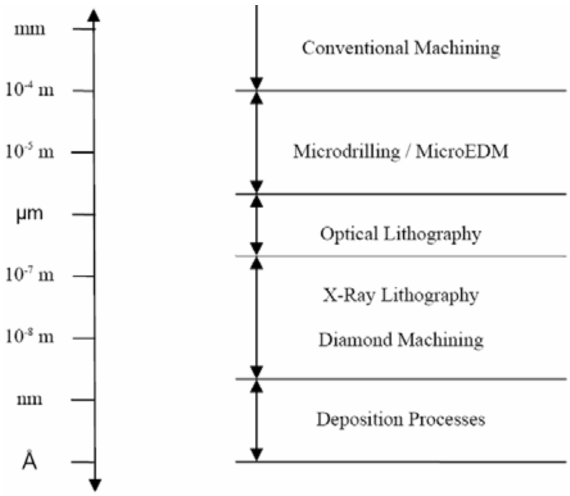
Intro
Fluid properties
Fluid dynamics
Heat transfer
Mass transfer
Chemical reactions
Conclusions

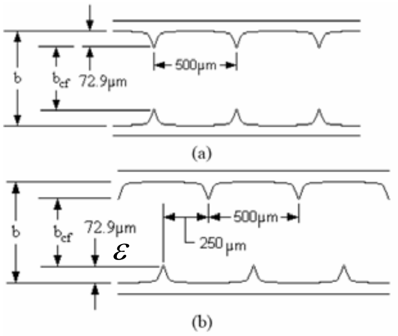
Surface roughness or texture

- Fabrication technologies
  - silicon etching
  - isotropic etching
  - mechanical fabrication



- feasible surface roughness





$$D_{cf} = d_h - 2\varepsilon \qquad \text{or}$$

$$D_{cf} = d_h - \varepsilon_1 - \varepsilon_2$$

J.B. Taylor et al., 2005

Transition regime

- turbulent flow regime
  transition depends on
  - surface roughness
  - inlet conditions

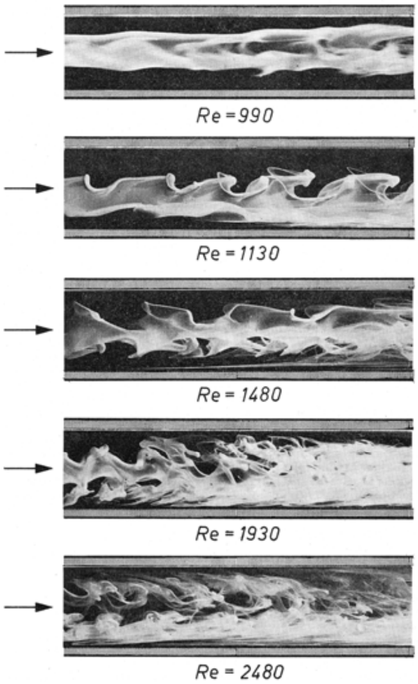
- critical Reynolds number

$$\mathrm{Re}_{\mathrm{krit}} = \frac{w_{\mathrm{krit}} d_h}{\nu} \approx 2300$$

- in microchannels
  - high flow velocities
  - high pressure loss
  - dissipative heating

- flow in bends and curves
  - first vortices at Re ≈ 10
  - secondary vortices at higher Re numbers
  - transient fluctuations at Re ≈ 200 - 400

Streak lines in tubular flow in transition regime laminar-turbulent



W. Albring, 1988

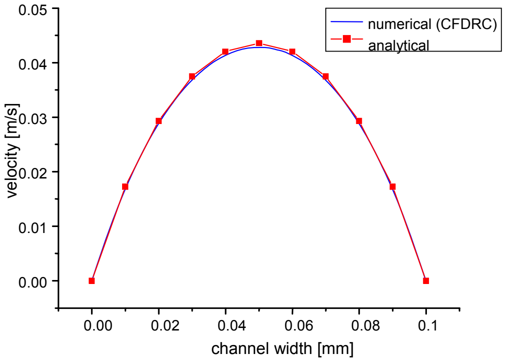
Analytical solutions

Intro
Fluid properties
Fluid dynamics
Heat transfer
Mass transfer
Chemical reactions
Conclusions

- Hagen- Poiseuille, capillary flow

$$\frac{\Delta p}{\Delta L} = \frac{128\,\eta}{\pi\, d_h^4}\dot{V}$$

$$w(r) = \frac{\Delta p}{\Delta L}\frac{D^2}{4\eta}\left[\left(\frac{r}{D}\right)^2 - \frac{1}{4}\right]$$



- flow in rectangular channel
  Fourier series or Prandtl's membrane analogy

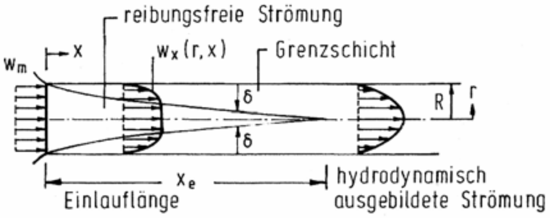
$$w(x,y) = \frac{\Delta p}{\eta L}\sum_{m,n}\frac{16\,b^2 h^2}{m\,n\,\pi^4\left(n^2 b^2 + m^2 h^2\right)}\sin\left(\frac{m\pi}{b}x\right)\sin\left(\frac{n\pi}{h}y\right)$$

- entrance flow

$$\frac{L_{in}}{d_h} = 0.056\ \mathrm{Re} \quad \text{or}$$

$$\frac{L_{in}}{d_h} = \frac{C_1}{1 + C_2\,\mathrm{Re}/C_1} + C_2\,\mathrm{Re}$$

	C_1	C_2
tube :	1.2	0.224
channel :	0.89	0.164



Baehr/Stephan, Wärme- und Stoffübertragung, 2004

Laminar flow in straight channels

- pressure loss

$$\Delta p = p_1 - p_2 = \sum_i \left(\lambda_i \frac{L_i}{d_{h,i}} \right) \cdot \frac{\rho}{2} w_1^2$$

$$\lambda_i = \frac{C_f}{\text{Re}}$$

C_f = 64 ○

= 56.92 ☐

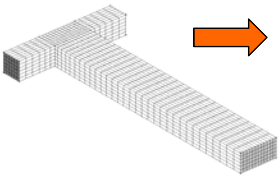
= 96 ▤

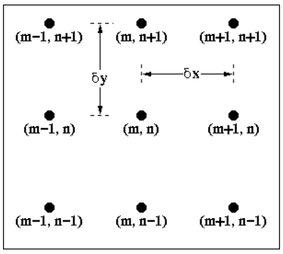
- channel cross section

Cross section, char. length		$C_f = \lambda_i$ Re	$w_{max}/\bar{w}$
circle, D		64	2.000
square, h		56.92	2.0962
rectangular, $h;b$ aspect ratio $\alpha_A = h/b$		$96\left[1 - 1.3553\alpha_A + 1.9467\alpha_A^2 - 1.7012\alpha_A^3 + 0.9564\alpha_A^4 - 0.2537\alpha_A^5\right]$	–
slab, $\alpha \to 0$		96	1.500
hexagon		60	–
60° trapezoid $h/b =$	4.00	55.66	2.181
	2.00	55.22	2.162
	1.00	56.60	2.119
	0.50	62.77	1.969
	0.25	72.20	1.766
KOH trapezoid h/b =1.00		56.15	2.137

Numerical solvers

- Finite Difference FD





$\delta x \quad x = m\delta x$

$\delta y \quad y = n\delta y$

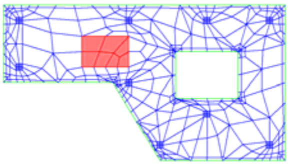
$$\frac{\partial f}{\partial x} \approx \frac{f(x_{m+1}) - f(x_{m-1})}{2\Delta x}$$

$$\frac{\partial^2 f}{\partial y^2} \approx \frac{f(x_m, y_{n+1}) - 2f(x_m, y_n) + f(x_m, y_{n-1})}{(\Delta y)^2}$$

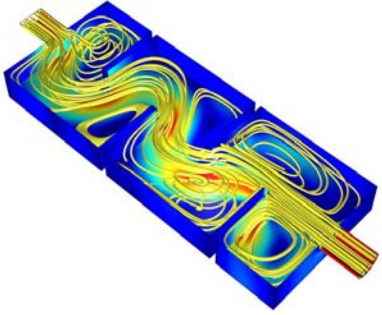
$$\nabla^2 f = \frac{\partial^2 f}{\partial x^2} + \frac{\partial^2 f}{\partial y^2}$$

$$\approx \frac{f(x_m, y_{n+1}) + f(x_m, y_{n-1}) + f(x_{m+1}, y_n) + f(x_{m-1}, y_n) - 4f(x_m, y_n)}{(\Delta x)^2}$$

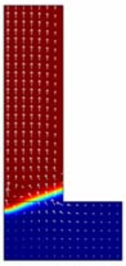
- FEM; z.B: COMSOL



Polynomial function in the element as solution function



flow and residence time in a reactor with baffles



Capillary filling of a microchannel

- Multiphysics Simulation

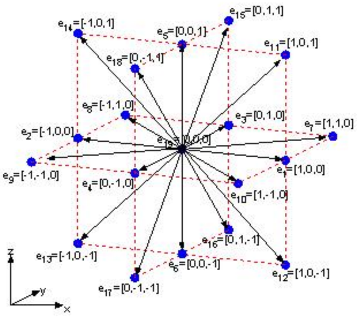
Numerical solvers

- Lattice Boltzmann methods LBM
  Cellular Automata

  approximation of the continuous
  Boltzmann equation on grid points

$$\frac{\partial f}{\partial t} + \vec{w} \cdot \frac{\partial f}{\partial \vec{x}} + \frac{1}{m} \; F \frac{\partial f}{\partial \vec{w}} = \frac{\sigma^2}{2} \int \left|\vec{V} \cdot \vec{e}\right| \left(f' f_1' - f \, f_1\right) \mathrm{d}\omega \, \mathrm{d}w_{x,1}$$

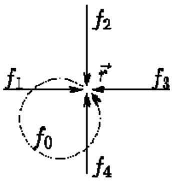
D3Q19: 3 dimensions, 19 points



www.latticeboltzmann.com/

D3Q27: 3 dimensions, 27 points

- formulation of collision term



$$f_i(\vec{r} + \lambda \vec{c}_i, t + \tau) - f_i(\vec{r}, t) = \sum_j \Omega_{ij} f_j(\vec{r}, t).$$

summation over
in- and outlet flows

Lab for Simulation

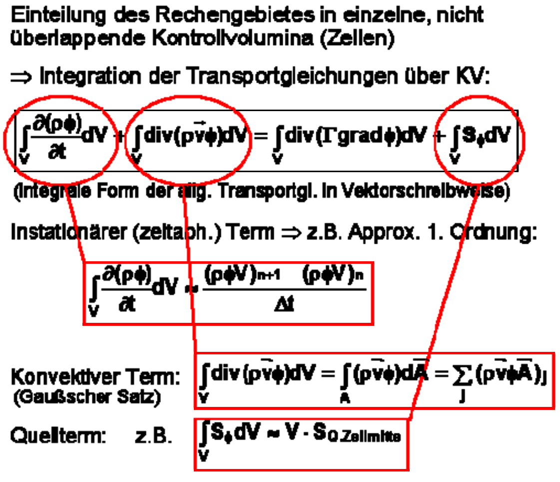
- Multiphysics simulation
  - turbulent flow and mixing
  - heat transfer
  - diffusion and chemical reactions

Chen, Doolen, 1998

www.imtek.de/simulation

Numerical solvers

- Finite Volume VOF: CFD-ACE+, Fluent, CFX, a.o.



Einteilung des Rechengebietes in einzelne, nicht überlappende Kontrollvolumina (Zellen)

⇒ Integration der Transportgleichungen über KV:

$$\int_V \frac{\partial(\rho\phi)}{\partial t} dV + \int_V \mathrm{div}(\rho\vec{v}\phi) dV = \int_V \mathrm{div}(\Gamma \,\mathrm{grad}\,\phi) dV + \int_V S_\phi dV$$

(Integrale Form der allg. Transportgl. in Vektorschreibweise)

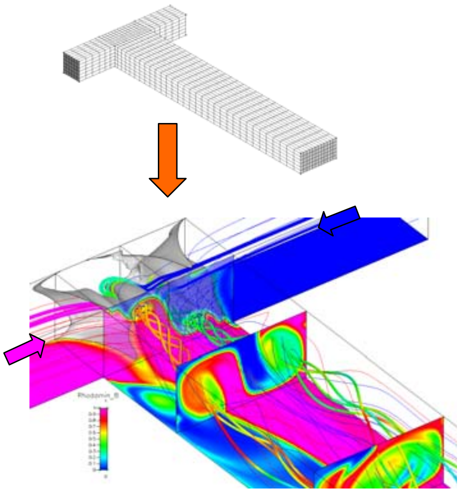
Instationärer (zeitabh.) Term ⇒ z.B. Approx. 1. Ordnung:

$$\int_V \frac{\partial(\rho\phi)}{\partial t} dV \approx \frac{(\rho\phi V)_{n+1} \quad (\rho\phi V)_n}{\Delta t}$$

Konvektiver Term: (Gaußscher Satz) $\int_V \mathrm{div}(\rho\vec{v}\phi) dV = \int_A (\rho\vec{v}\phi) d\vec{A} = \sum_j (\rho\vec{v}\phi\vec{A})_j$

Quellterm: z.B. $\int_V S_\phi dV \approx V \cdot S_{\phi,\mathrm{Zellmitte}}$

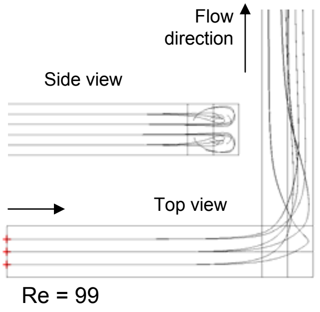
www-ifkm.mach.uni-karlsruhe.de

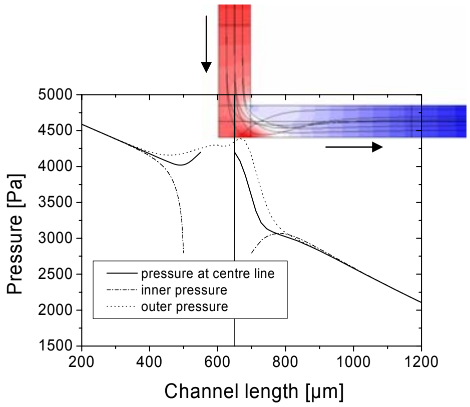


- Pro‘s and Con‘s of numerical solutions:
  - **FD**, “exact“ approximation, rectangular lattice,
  - **VOF**, “exact“ approximation, various elements,
  - **LBM**, rapid method with arbitrary geometries,
  - **FEM**, multiphysics, solution depends on applied polynom.

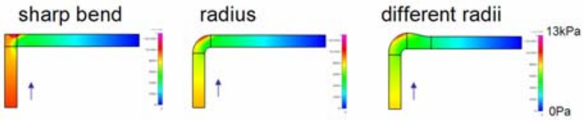
Channel structures, bends

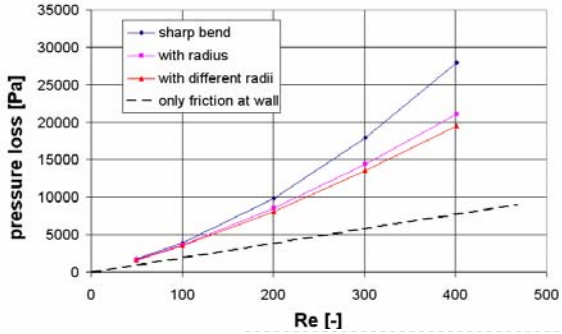
- laminar flow in curves and bends
  Dean flow
  generation of vortex pairs





- pressure loss





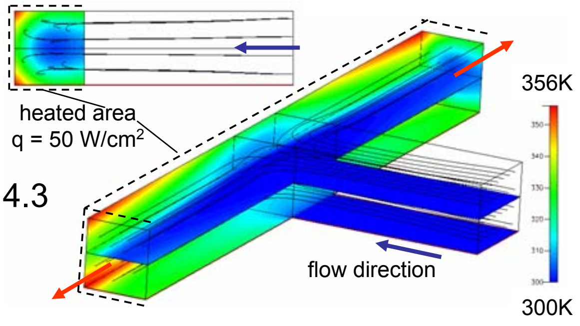
$$\Delta p = p_1 - p_2 = \sum_i \left(\lambda_i \frac{L_i}{d_{h,i}} + \zeta_i \right) \cdot \frac{\rho}{2} w_1^2$$

$$\Rightarrow \Delta p = m^* \frac{\rho}{2} w_1^n = m \mathrm{Re}^n; \quad n = 1.2 - 1.8$$

Heat Transfer – Laminar Flow

- Simulation of the heat transfer in a micro channel:
  - Constant wall heat flux, 50 W/cm²,
  - Single phase flow, water,
  - Definition of the Nusselt number:

$$\mathrm{Nu}_q = \frac{h_q d_h}{k} = \frac{q d_h}{k\left(\overline{T}_W - \overline{T}_F\right)}$$



- Laminar developed flow:

$q = const.$ → Nu = $const.$ = 4.3

- Entrance flow and
  bend flow lead to vortices:
  → Nu number increases.

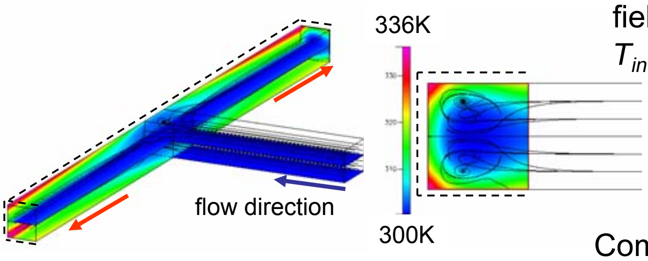
T_{in} = 300K, water, Re_{in} = 26
d_h = 100µm, water

$h_q \approx 25.8\ \mathrm{kW/m^2K}$

Intro
Fluid properties
Fluid dynamics
Heat transfer
Mass transfer
Chemical reactions
Conclusions

Heat Transfer – Curved Flow

- Heat transfer in a T-joint micro channel with constant heat flux:



Streamlines and temperature fields, $Re_{in} = 155$, $Re_{out} = 109$, $T_{in} = 300$ K

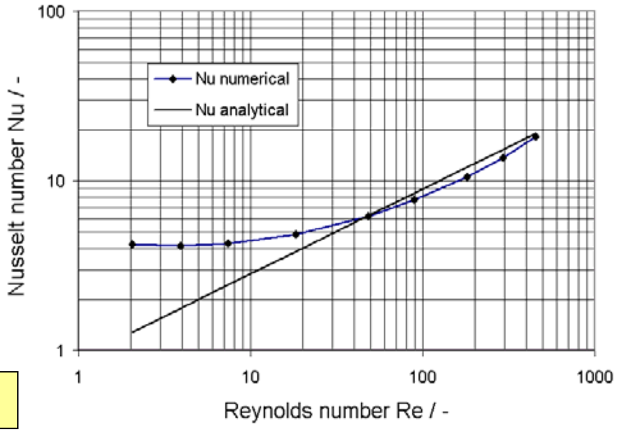
Heat transfer enhancement due to vortices.

Analytical calculation for developing flow consitions:

$$\mathrm{Nu}_q = 0.664\,\mathrm{Pr}^{1/3}\sqrt{\mathrm{Re}\frac{d_h}{L}}$$

$$\mathrm{Nu}_q \approx 20 \rightarrow h_q \approx 120\ \mathrm{kW/m^2K}$$

Comparison simulation - theory



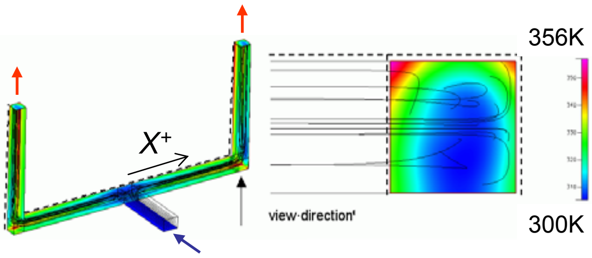
Intro
Fluid properties
Fluid dynamics
Heat transfer
Mass transfer
Chemical reactions
Conclusions

Heat Transfer – Curved Flow

- Microchannels with bends and joints:
  - Entrance flow at each bend leads to vortices:
    → Nu number is increased.
  - Dimensionless length X^+

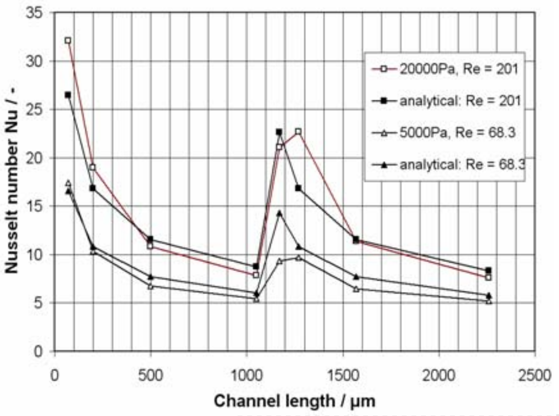
$$X^+ = \frac{L}{d_h \mathrm{Pe}} = \frac{L}{d_h \mathrm{Re}\cdot\mathrm{Pr}}$$

  - Mean Nu number for the channel



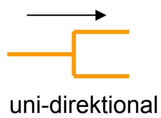
$$\mathrm{Nu}_{me} = \frac{\mathrm{Nu}_m}{\tanh\left(2.432\mathrm{Pr}^{1/6}\,X^{+1/6}\right)}$$

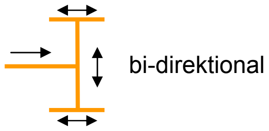
- Good agreement between simulation and analytical results.



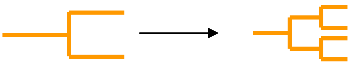
Channel networks

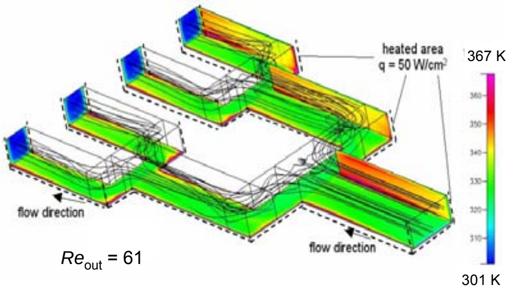
- Layout acc. to Bejan‘s „*constructal*“ design method:
  - element of 0th. order
  - systems setup

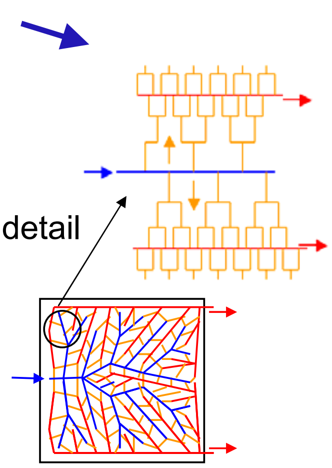


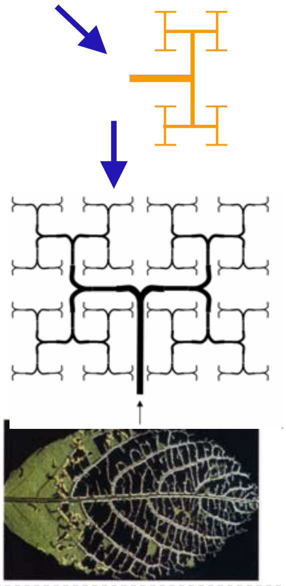


- design of complex structures









- but: high pressure loss

Cross sections in channel systems

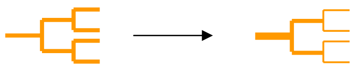
- Biological structures follow a certain pattern
- Murray‘s Law (also W.R. Hess, 1914)

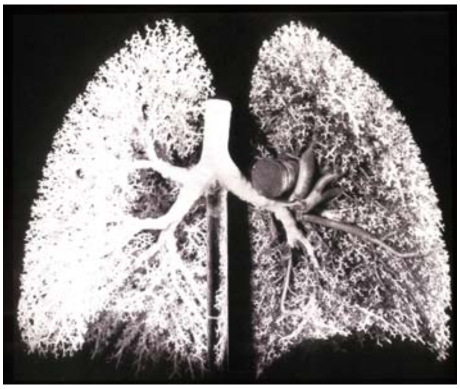
- Example: Fork-shaped structure

1. Level z-Level

$$d_{h,0}^3 = \sum_i d_{h,i}^3$$

$$d_{h,1} = d_{h,0}\, 2^{-1/3}; \quad d_{h,z} = d_{h,0}\, 2^{-z/3}$$



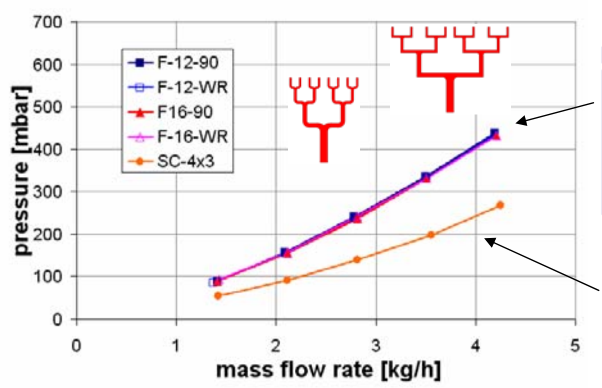


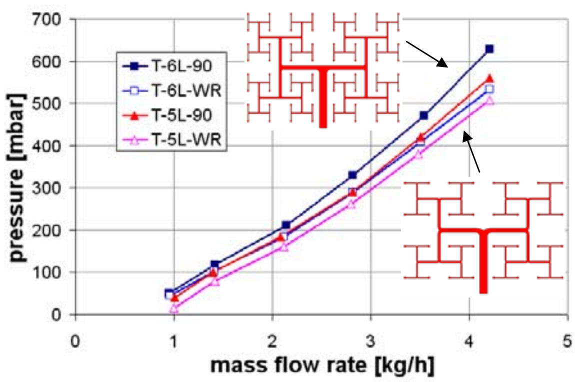
Lungenflügel, www.uni-ulm.de/ klinik/chirurgie2/ images/SPL-009.jpg

- Constant wall shear stresses τ_W
- Biggest resistance at the smallest parts
- Stabilized flow and homogeneous flow distribution

Intro
Fluid properties
Fluid dynamics
Heat transfer
Mass transfer
Chemical reactions
Conclusions

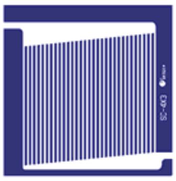
Channel networks: Pressure loss





Fork-shaped device

- No influence of single branches



Conventional device

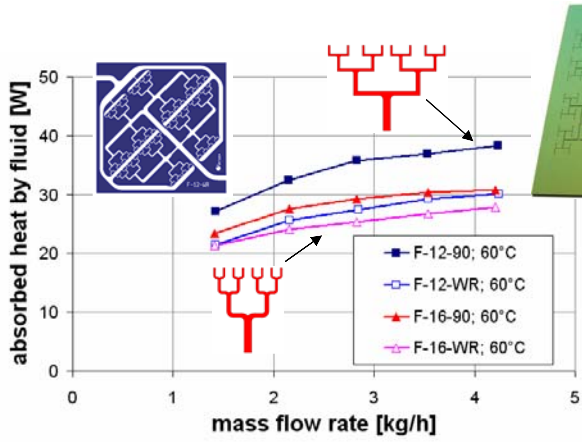
- Half of the pressure loss

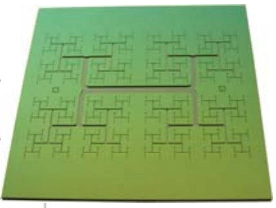
T- tree device

- Has the highest pressure loss
- Wedges reduce the pressure loss by 10-15%

Intro
Fluid properties
Fluid dynamics
Heat transfer
Mass transfer
Chemical reactions
Conclusions

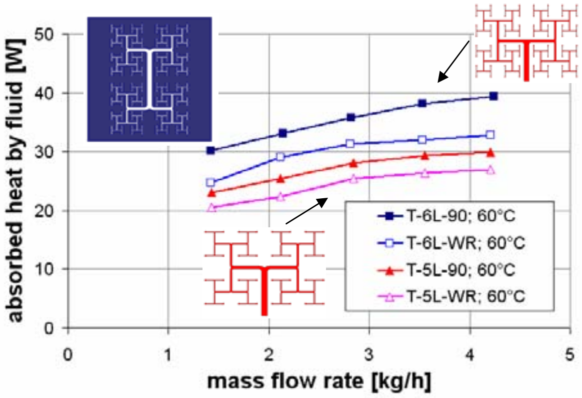
Channel networks: Heat transfer





Fork-shaped device

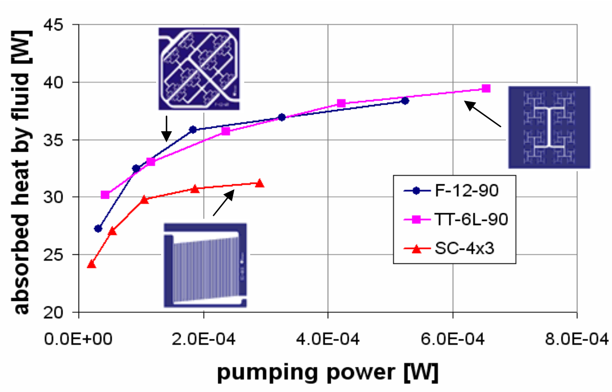
- Wider branches have a higher heat transfer rate
- Pressure optimized versions reduce the heat transfer significantly



- **T- tree device**
- More levels increase the heat transfer significantly
- Pressure optimized versions reduce the heat transfer significantly

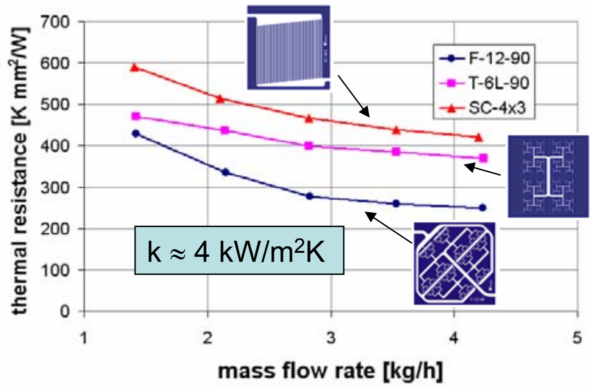
Intro
Fluid properties
Fluid dynamics
Heat transfer
Mass transfer
Chemical reactions
Conclusions

Experimental results: Comparison



Absorbed heat

- 18% higher heat transfer rate at equal pumping power
- 26% higher heat transfer rate at equal mass flow rate



Thermal resistance

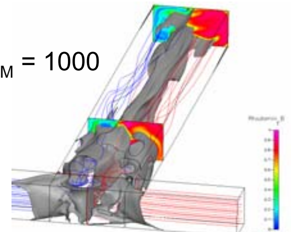
$$R_{th} = \frac{\left(\overline{T}_{chip} - \overline{T}_{fluid}\right) \cdot A_{chip}}{\dot{Q}}$$

- 38% lower thermal resistance at equal pumping power
- 41% lower thermal resistance at equal mass flow rates

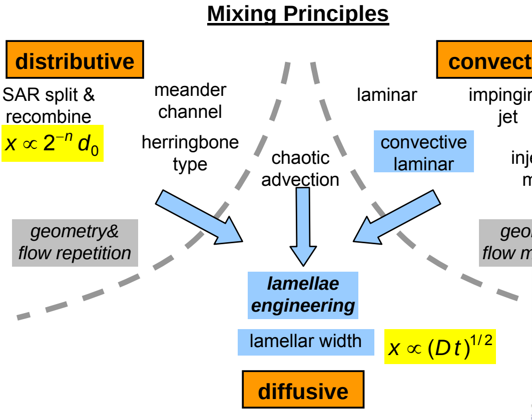
Intro
Fluid properties
Fluid dynamics
Heat transfer
Mass transfer
Chemical reactions
Conclusions

Overview mixing principles

- Mixing in microfluidics by advection, convection, and diffusion



Re_M = 1000



Mixing Principles

distributive

Interdigital

Focus mixer

SAR split & recombine

$x \propto 2^{-n} d_0$

meander channel

herringbone type

geometry& flow repetition

chaotic advection

laminar

convective

impinging jet

turbulent

$x \propto (\nu / \varepsilon)^{1/4}$

convective laminar

injection mixer

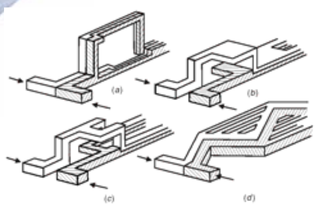
geometry & flow momentum

lamellae engineering

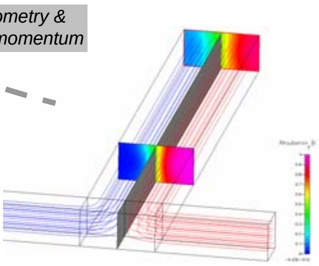
lamellar width

$x \propto (D t)^{1/2}$

diffusive



acc. Nguyen, Wu, 2005

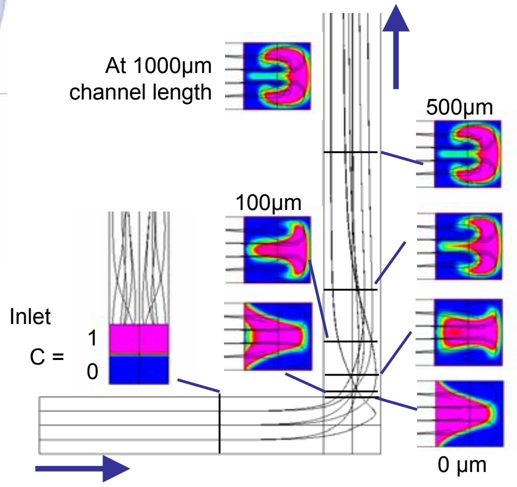


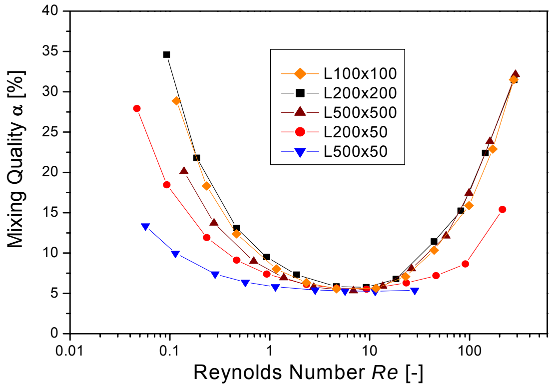
Re_M = 0.01

- Main goal: reduction of diffusion length
- Control of striation thickness

Mixing in sharp 90° bend

- Simulation: streamlines and concentration fields
  - 90° sharp bend (100×100 µm²)
  - Re = 99, *w* = 0,85 m/s
  - Re > 10 → vortex generation





residence time: $t_P = \frac{l}{\bar{u}}$

diffusion time: $t_D = \frac{(d/2)^2}{2D} = \frac{d^2}{8D}$

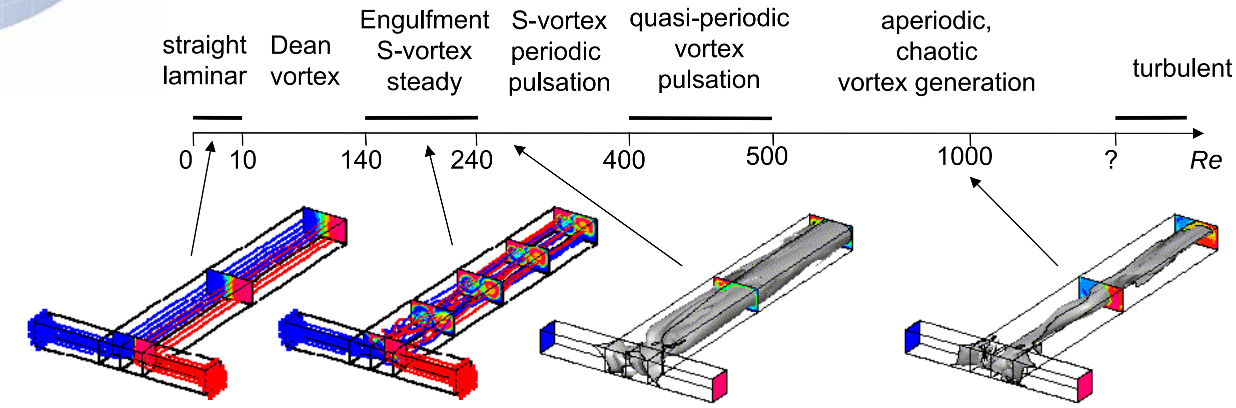
$$\frac{t_D}{t_P} = \frac{d}{8l}\mathrm{Re}\cdot\mathrm{Sc} = \frac{\mathrm{Pe}}{8}\frac{d}{l} \qquad \alpha \propto \frac{1}{\mathrm{Re}\,\mathrm{Sc}} = \frac{1}{\mathrm{Pe}}$$

Dean number $\mathrm{Dn} = \mathrm{Re}\left(\frac{d_h}{2R}\right)^{1/2}$

Schmidt number $\mathrm{Sc} = \frac{\nu}{D}$

Channel structures, T-mixer

- transient flow in T-shaped micromixers
  generation of vortex pairs and vortex shedding, wake flow
- symmetrical mixing 1:1
- asymmetrical flow for Re > 140
- periodic vortex pulsation for 240 < *Re* < 400
- quasi-periodic pulsation for 400 < *Re* < 500
- chaotic pulsations for 500 < *Re* < 1000 (min.)



straight laminar

Dean vortex

Engulfment S-vortex steady

S-vortex periodic pulsation

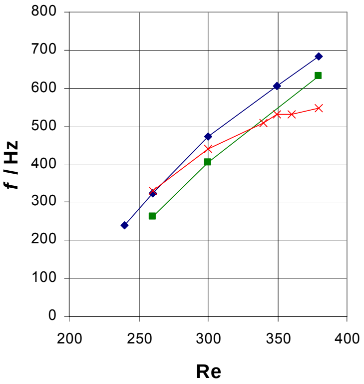
quasi-periodic vortex pulsation

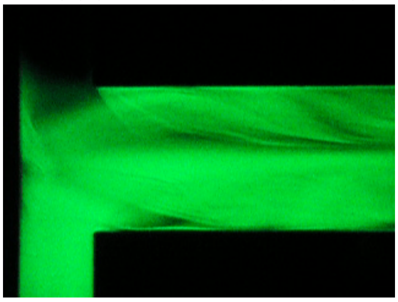
aperiodic, chaotic vortex generation

turbulent

Transient flow regimes

- transient flow regimes in T-mixers, rectangular cross section
  typical frequencies ~ 500 Hz for T600x300x300, d_h = 400µm
  ~ 4500 Hz for T200x100x100, d_h = 133µm



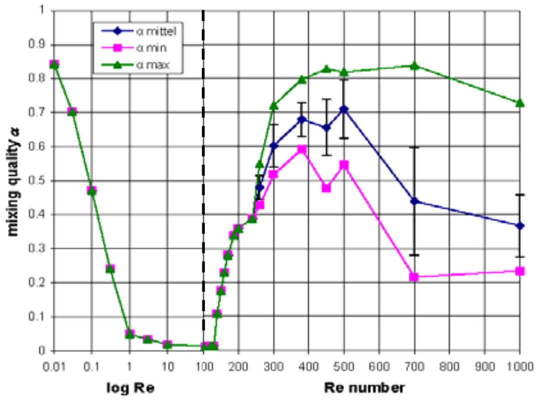


Re = 300, channel depth 300 µm
mixing channel width 600 µm
f = 440 Hz
fluorescence color Uranin

$$\text{Strouhal-Zahl Sr} = \frac{f \cdot d_h}{w} \approx 0.2$$

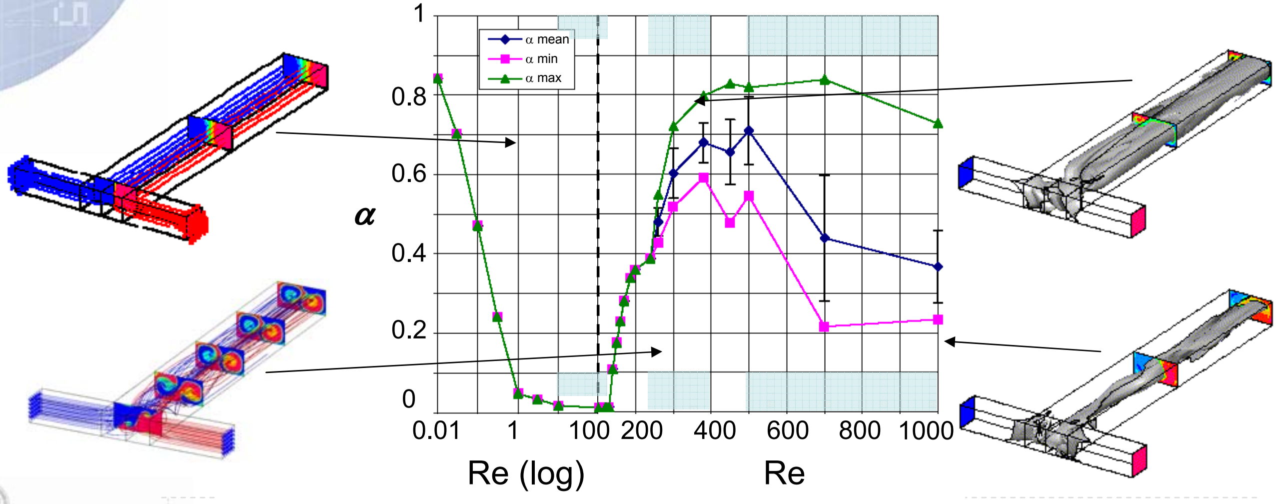
- Influence on mixing
  Mixing quality

$$\alpha = 1 - \sqrt{\sigma^2 / \sigma^2_{max}}$$



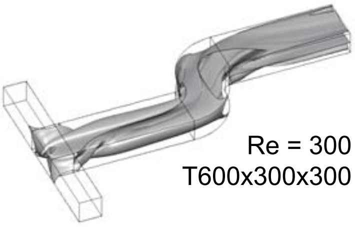
Mixing regimes: Influence on the mixing quality

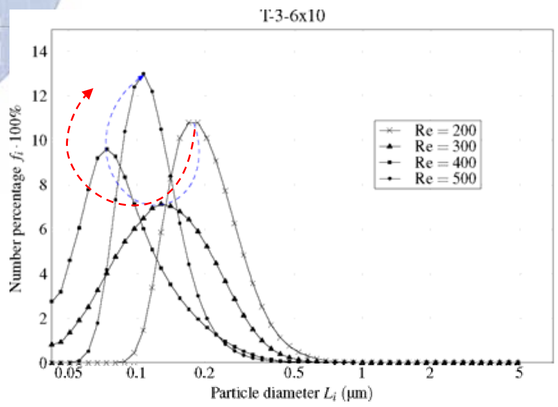
- Mixing quality (after 2 mm) depending on *Re*:
  - straight laminar und vortex flow:
    residence time is important
  - Engulfment and periodic pulsation: lamellae generation
  - chaotic flow: separation interface with bursts

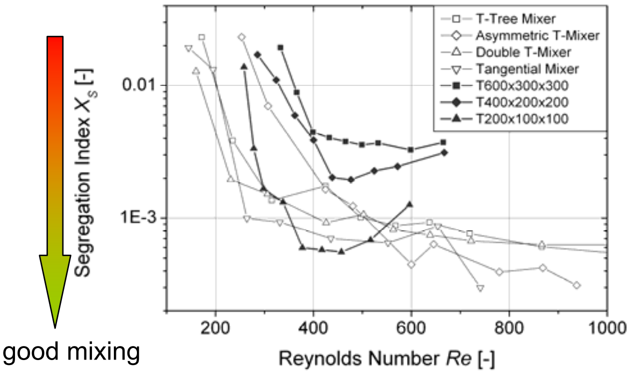


Results from chemical reactions

- Mixing in T-shaped device
- unsteady flow behavior in micromixers
- mixing in convective mixers
- particle precipitation $BaSO_4$
- parallel-competitive reaction


Re = 300
T600x300x300

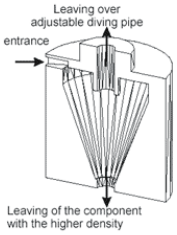


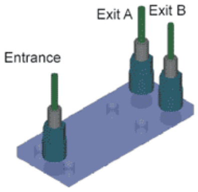


➡comparable behavior in precipitation and chemical reaction
➡advanced devices necessary

Channel structures, mixer

- Cyclone

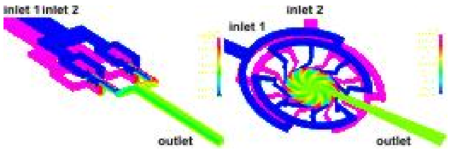


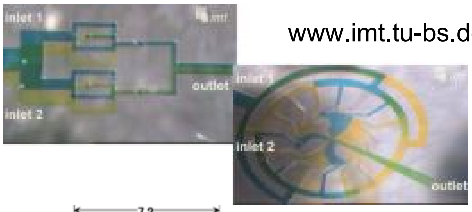


www.ltf-gmbh.de


cyclone insert, FZK, Germany

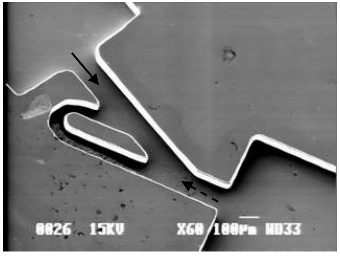
- Vortex mixer

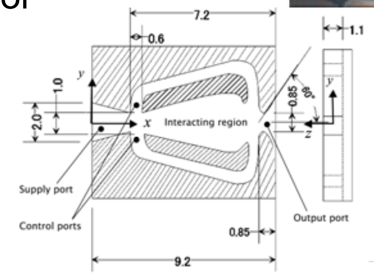




www.imt.tu-bs.de

- Tesla valve, fluidic oszillator





Channel structures, axial dispersion

- mean residence time, plug flow

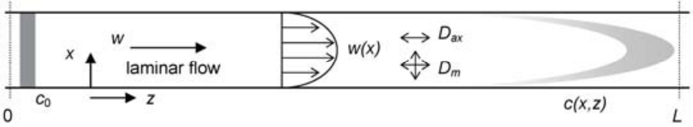
$$t_P = L / \overline{w}$$

- axial dispersion by diffusion and laminar convection

$$J_n = -D_{ax} A \frac{\partial c}{\partial z} \qquad \frac{\partial c}{\partial t} = -w \frac{\partial c}{\partial z} + D_{ax} \frac{\partial^2 c}{\partial z^2}$$

$$D_{ax} = D_m + \frac{w^2 d_h^2}{C_D \cdot D_m}$$

$$C_D \approx 192 - 210$$

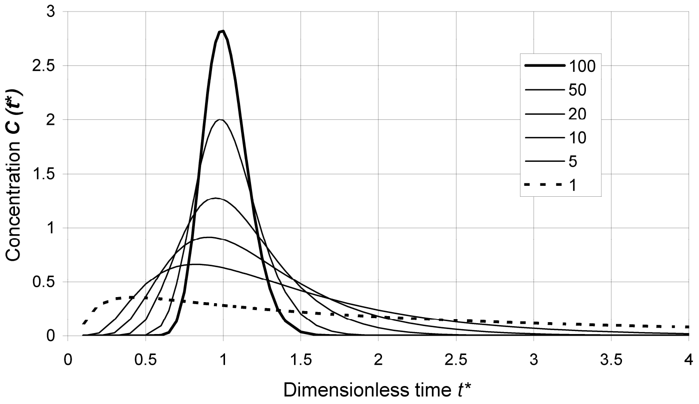


- Bodenstein number

$$\mathrm{Bo} = \frac{w \cdot L}{D_{ax}}$$

$$\frac{\partial c^*}{\partial t^*} = -\frac{\partial c^*}{\partial z^*} + \mathrm{Bo} \frac{\partial^2 c^*}{\partial z^{*2}}$$

- residence time distribution

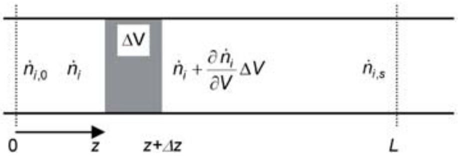


Chemical reactions in continuous flow

- reaction rate

$$\dot{n}_i - \dot{n}_{i,0} = V \cdot R_i = V \rho_m \sum_j \nu_{ij} \cdot r_j$$

with $r_j = \pm k_j \prod_i c_i^{m_i}$

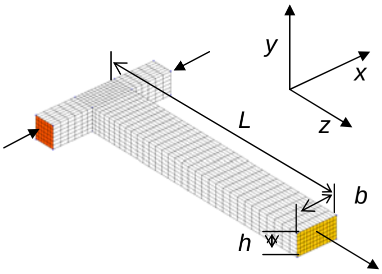


- chemical reaction with diffusion and convection

$$\frac{\partial c}{\partial t} = -w \frac{\partial c}{\partial z} + D_{ax} \frac{\partial^2 c}{\partial z^2} + \sum_j \nu_{ij} \cdot r_j$$

- typical time scales
  - reaction time $t_R = \frac{c_i}{\upsilon_i \, r} \sim 1/k$
  - residence time $t_P = \frac{L_R}{\overline{w}}$
  - diffusive mixing time $t_D = \frac{(b/2)^2}{2D_m} = \frac{b^2}{8D_m}$

  „turbulent“ micro mixing $t_E = 17.3 \left(\frac{\nu}{\varepsilon} \right)^{1/2}$



- time scale ratio is expressed by Damköhler numbers

Intro
Fluid properties
Fluid dynamics
Heat transfer
Mass transfer
Chemical reactions
Conclusions

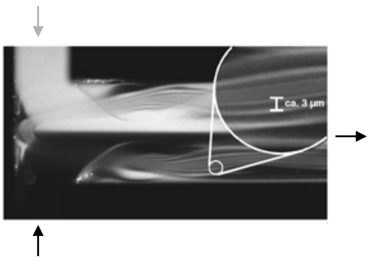
Characteristic times of reaction and transport

- reactions and mass transfer
  - residence time $\frac{t_P}{t_R} = \frac{\nu_i \, r \, L}{c_i \, \overline{w}} = \mathrm{DaI}$
  - diffusive mixing time $\frac{t_m}{t_R} = \frac{\nu_i \, r \, d_h^2}{c_i \, D_m} = \mathrm{DaII}$
- design criteria
  DaI > 1 complete reaction
  DaII < 1 fast mixing, „pre-mixed“ reaction
- reaction heat ΔH_r and heat transfer
  - heat capacity of fluids $\frac{\rho_m \, \Delta H_r \, r \, L}{\rho \, c_p \, \Delta T_z \, \overline{w}} = \mathrm{DaIII}$
  - convective heat transfer $\frac{\rho_m \, \Delta H_r \, r \, d_h^2}{\lambda_w \, \Delta T_W} = \mathrm{DaIV}$
- design criteria
  DaIII ~ 1 isothermal reaction
  DaIV < 1 mitigation of hot spots

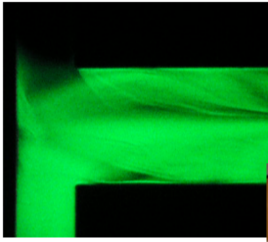
Intro
Fluid properties
Fluid dynamics
Heat transfer
Mass transfer
Chemical reactions
Conclusions

Summary

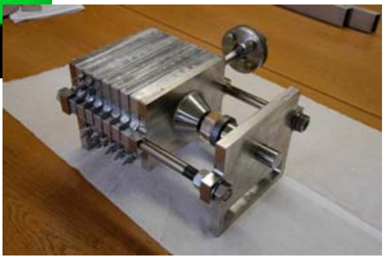
- Why micro process engineering?



- Scaling and fluid behavior
- Laminar flow regime and vortex generation



- Channel elements



- Microfluidic devices with chemical reactions

Intro
Fluid properties
Fluid dynamics
Heat transfer
Mass transfer
Chemical reactions
Conclusions

Thanks to

Many of the presented results have been funded and supported by the following organizations:

Funding by the **DFG** (German Research Foundation) in the

Priority program SPP1141 „Strömungsmischer“

Research project „Effective micromixer“

Deutsche Forschungsgemeinschaft DFG

Funding by Country Baden-Württemberg

„Integrated Processes with Microreactors“

Lonza AG, Visp, R&D Exclusive Synthesis

Thank You for Your Attention!

Literature

Textbooks and Monographs

- G.K. Batchelor, An Introduction to Fluid Dynamics, Cambridge University Press, 2000
- P. Tabeling, Introduction to Microfluidics, Oxford University Press, 2005
- N. Kockmann, Transport Phenomena in Micro Process Engineering, Springer, 2007
- N. Kockmann (Ed.), Micro Process Engineering, Wiley-VCH, 2006
- J.J. Brandner et al., Microfabrication in Metals and Polymers, Chap. 10 in Micro Process Engineering, N. Kockmann (Ed.), Wiley-VCH, 2006
- O. Tonomura, Simulation and analytical modeling for microreactor design, Chap. 8 in Micro Process Engineering, N. Kockmann (Ed.), Wiley-VCH, 2006
- F. Goldschmidtböing et al., Silicon microfabrication for microfluidics, Chap. 11 in Micro Process Engineering, N. Kockmann (Ed.), Wiley-VCH, 2006
- Y. Sone, Kinetic theory and fluid dynamics, Birkhäuser, 2002
- C. Cercignani, The Boltzmann equation and its applications, Springer, 1988
- M. Gad-el-Hak, The MEMS Handbook, CRC Press, 2006
- A. Bejan, Shape and Structure, from Engineering to Nature, Cambridge University Press, 2000
- H.D. Baehr, K. Stephan, Wärme- und Stoffübertragung, Springer, 2004
- W. Albring, Angewandte Strömungslehre, Akademie-Verlag, 1988

Literature

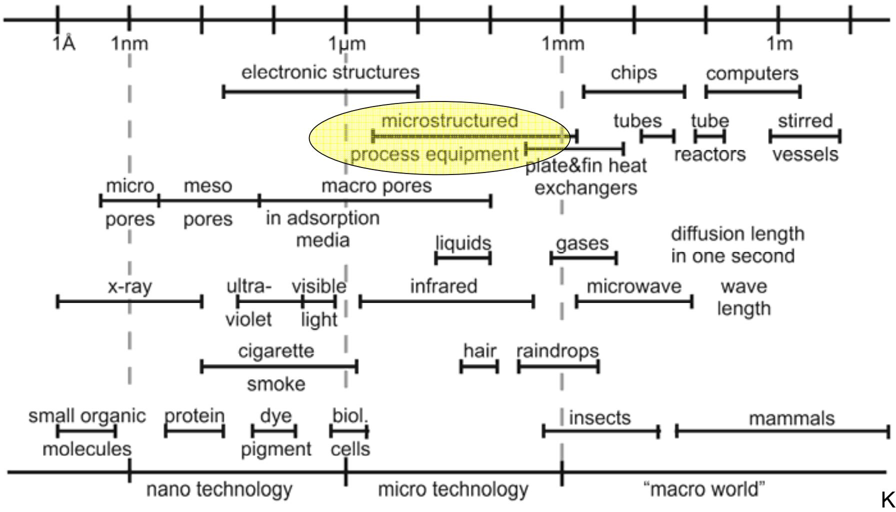
Journal and conference papers

- M. Schlüter, M. Hoffmann, N. Räbiger, Theoretische und experimentelle Untersuchungen der Mischvorgänge in T-förmigen Mikroreaktoren – 2: Experimentelle Untersuchungen des Strömungsmischens, Chemie-Ingenieur-Technik 76 (2004) 1682-1688
- N. Kockmann, M. Engler, P. Woias, Theoretische und experimentelle Untersuchungen der Mischvorgänge in T-förmigen Mikroreaktoren – 3: Konvektives Mischen und chemische Reaktionen, Chemie-Ingenieur-Technik 76 (2004) 1777-1783
- S. Nedea et al. Density distribution for a dense hard-sphere gas in micro/nano-channels: Analytical and simulation results, J. Comp. Physics 219 (2006) 532–552
- R.W. Barber, D.R. Emerson, Challenges in Modeling Gas-Phase Flow in Microchannels: From Slip to Transition, ICNMM2005-75074, 2005
- J.B. Taylor, A.L. Carrano, S.G. Kandlikar, Characterization of the Effect of Surface Roughness and Texture on Fluid, ICNMM2005-75075, 2005
- L. Cheng, D. Mewes, Review of two-phase flow and flow boiling of mixtures in small and mini channels, Int. J. Multiphase Flow 32 (2006) 183-207
- E.V. Rebrov et al. Header Design for Flow Equalization in Microstructured Reactors, AIChE J. 53 (2007) 28-38
- S. Chen, G.D. Doolen, Lattice Boltzmann Method for Fluid Flows, Annu. Rev. Fluid Mech. 30 (1998) 329-364
- G. Hetsroni, A. Mosyak, E. Pogrebnyak, L.P. Yarin, Fluid flow in microchannels, Int. J. Heat Mass Transfer, 48 (2005) 1982-1998
- D.J. Quinn et al. A Systematic Approach to Process Selection in MEMS, J. MEMS 15 (2006) 1039-1050

Length scales

Intro
Fluid properties
Fluid dynamics
Heat transfer
Mass transfer
Chemical reactions
Conclusions

- typical regimes



Kockmann 2006

- micro structures enlarge the process space
- on small length scales, the behavior of single atoms or molecules becomes important
- fluid properties may change!

Typical dimensions, where pulsations do occur?

- periodic fluctuations at 240 < Re < 400
- typical frequencies of 500 Hz, Sr ≈ 0,2
- typical channel dimensions 100 < d_h < 1000 µm
  for water at 20 °C