CHISA 2014
PRAGUE PRES 2014

# Particle migration effects as a basis for effective microfiltration

August 27, 2014: Karin Schroën, Anna van Dinther, Remko Boom

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# Overview

- Fractionation / separation of components

  Dairy as an example: principle mechanisms

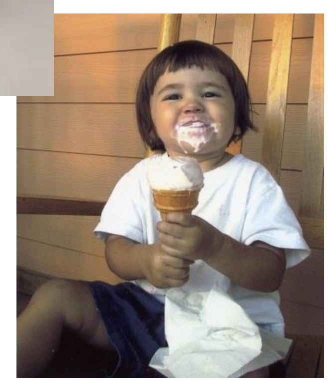
- Particle migration phenomena
  - Simulations
  - CSLM
  - Membranes (low, high concentration, actual food)

- Process design

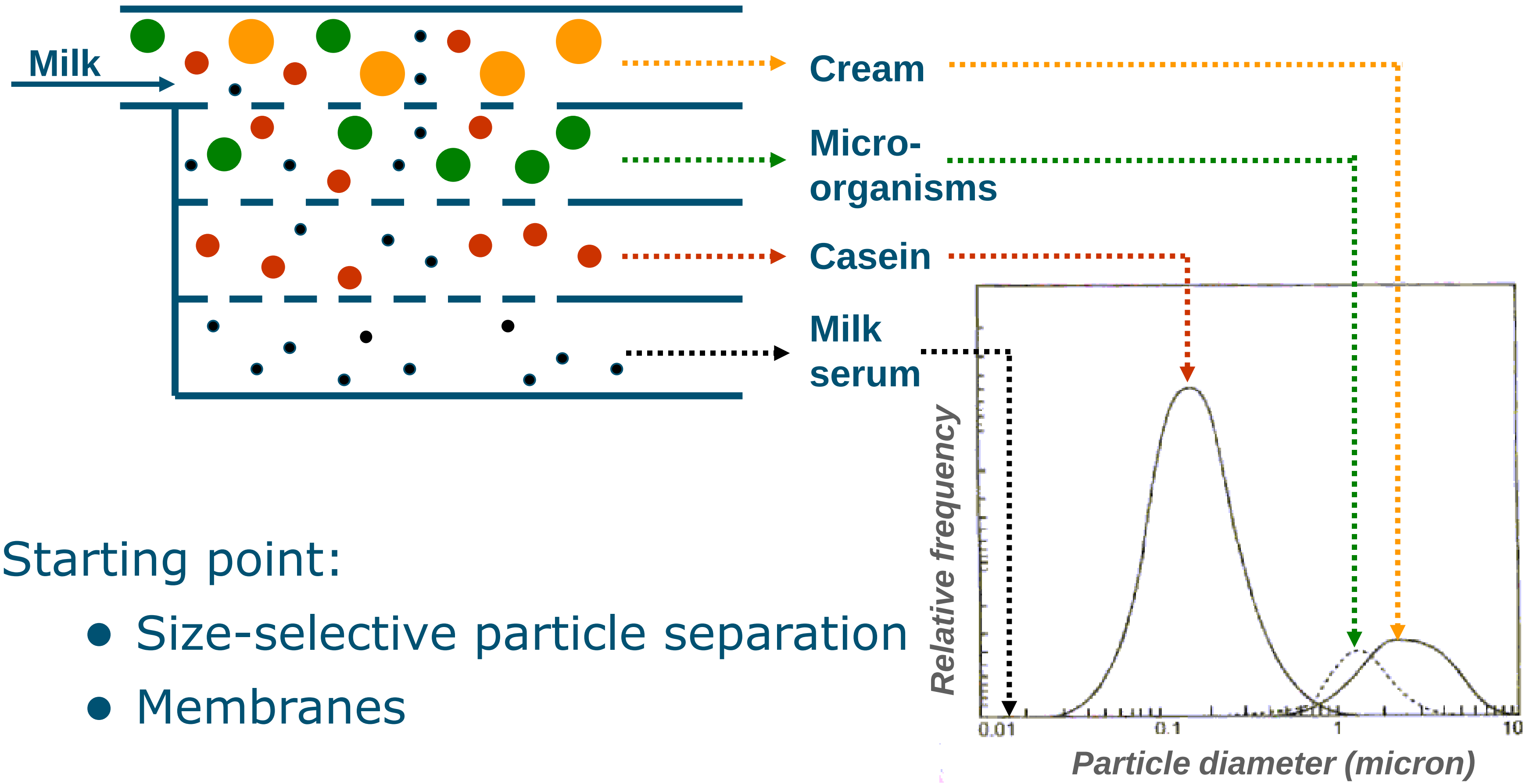
# Milk fractionation

- Increased shelf life
- ‘Fresher’ products (cold sterilisation)
- Less transport costs
- Better defined starting materials
- New products


➢ Other processes

# Fractionation of milk

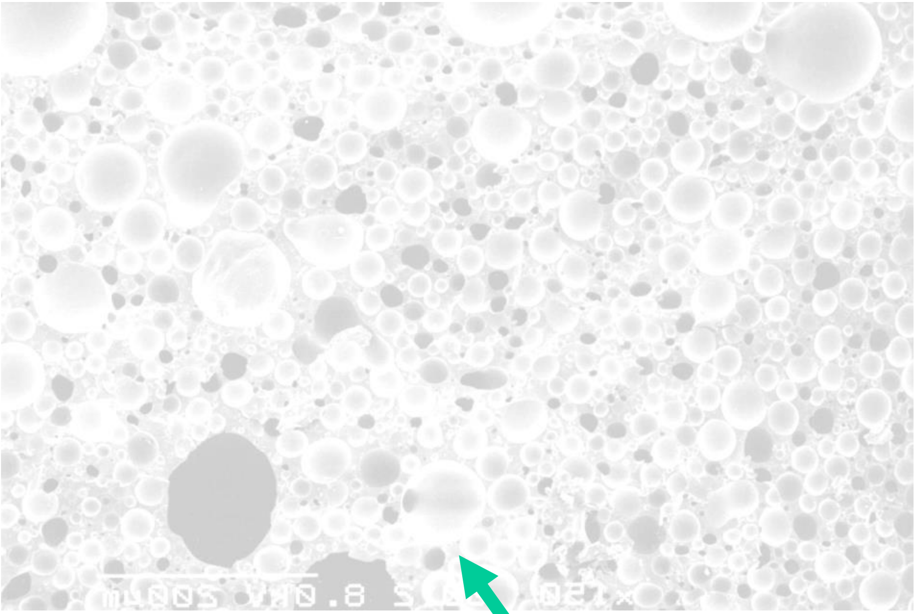


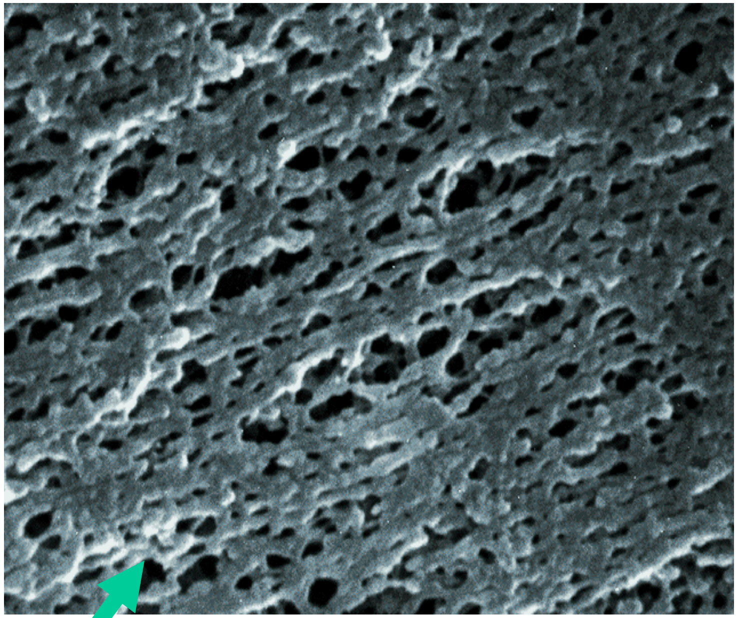

- Starting point:
  - Size-selective particle separation
  - Membranes



*Brans et al, Journal of Membrane Science (2004) 243, p263*

# Fractionation of milk





- So we are trying to separate this with this!

# Surprisingly: sometimes it works!

Cold sterilisation
Separation of milk fat from milk
Concentration of casein
Recovery of serum protein from whey

But not complete fractionation!

# Cold sterilisation

| Membrane type and flux | Process conditions cross flow / pressure, UTP, back pulsing | Log reduction | Source |
| --- | --- | --- | --- |
| Ceramic 1.4 µm; $1.4 \cdot 10^{-4}$ m/s | 50 kPa, 7.2 m/s UTP | above 3.5 | Saboya and Maubois 2000 Lait |
| Reversed asymmetric 0.87 µm; $1.4 \cdot 10^{-4}$ m/s | 0.5-1 m/s; back pulsing 0.2-1 $s^{-1}$ | between 4 and 5 | Guerra et al. 1997 Int. Dairy Journal, |
| Microsieve 0.5 µm | dead-end filtration of spiked SMUF | 6.6 | Van Rijn and Kromkamp (patent) |
| Bactocatch: ceramic membranes | 6 to 8 m/s | | Holm et al. (patent) |



*Schroën et al, 2010 Membrane Technology, Volume 3: Membranes for Food Applications*

# Concentration of casein

| Membrane type and flux | Process conditions cross flow / pressure | Concentration factor | Source |
|---|---|---|---|
| Ceraflo 0.22 µm; $2.5 \cdot 10^{-5}$ m/s | 6.9 m/s; 190 kPa | 3 | Pouliot et al. 1996 Int. Dairy J. |
| Membralox 0.2 µm $1.9 \cdot 10^{-5}$ - $1.3 \cdot 10^{-5}$ m/s | 7.2 m/s; 193 kPa | 2-10 | Vadi and Rizvi 2001 JMS |
| Ceramem asymmetric 0.05 µm; $3.1 \cdot 10^{-5}$ m/s | 5.4 m/s; 138 kPa | 2 | Punidadas and Rizvi 1999 Food Res. Int |
| Membralox 0.1 µm; $9.7 \cdot 10^{-5}$ - $2.5 \cdot 10^{-4}$ m/s | 0.45 m/s; 34 kPa turbulence promoters 12.5 m/s; 65 kPa | 1 | Krstic et al. 2002 JMS |



*Schroën et al, 2010 Membrane Technology, Volume 3: Membranes for Food Applications*

# Surprisingly: sometimes this works!

A lot of measures are taken to keep the process running

- Back pulsing / shocking (very frequent)
- Turbulence promoters
- Critical flux concept
- Uniform transmembrane pressure concept
- (acoustic waves, sonication)

➢ And lots of cleaning!

# Is there any rationale behind this?

'Any measure that gets rid of accumulated particles is good.'

But is this the 'best' approach?

- Focus on particles (starting point of our research)
- Focus on membrane and module design

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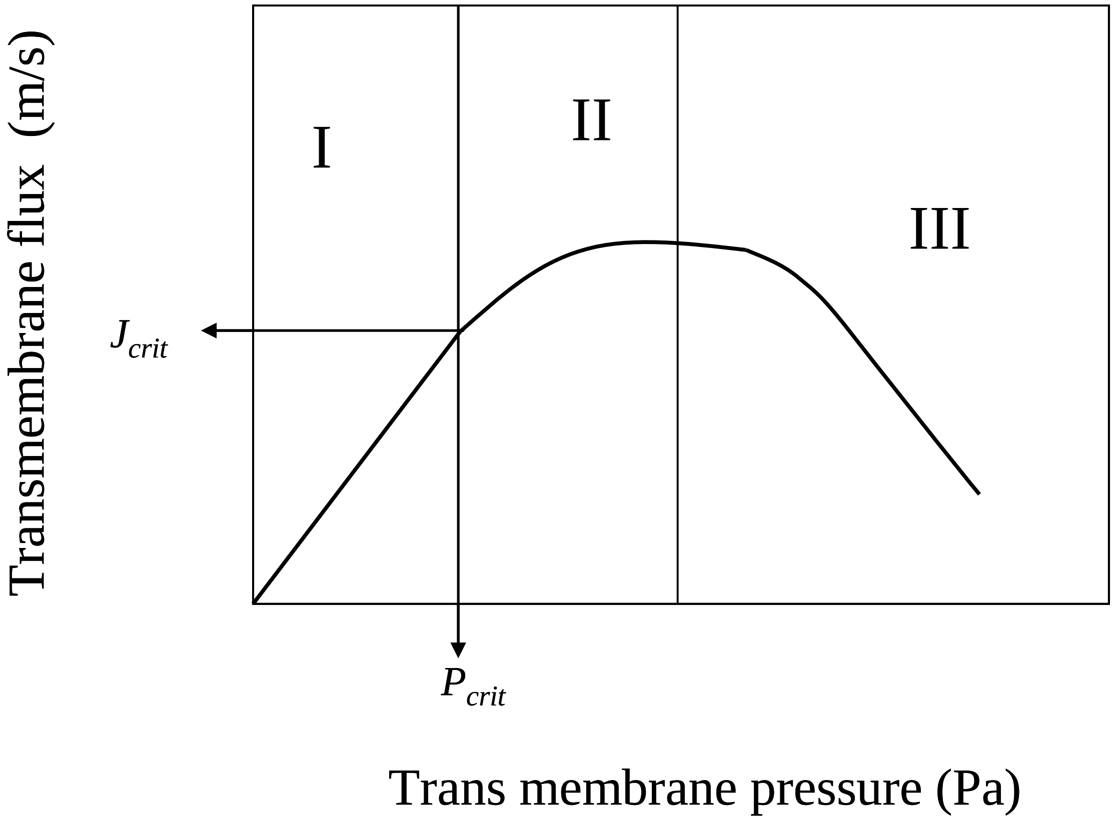
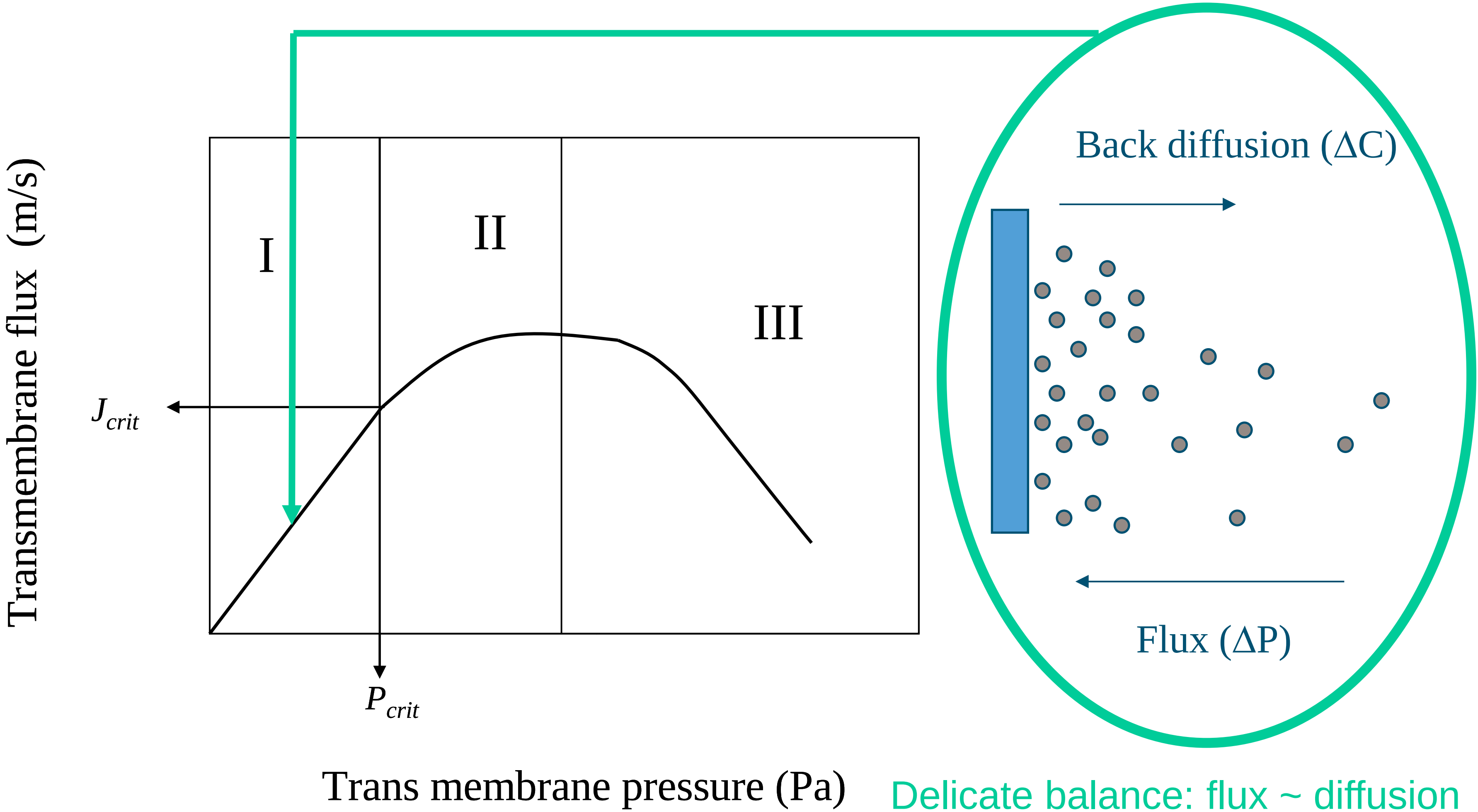
# Critical flux: starting point ‘particles’

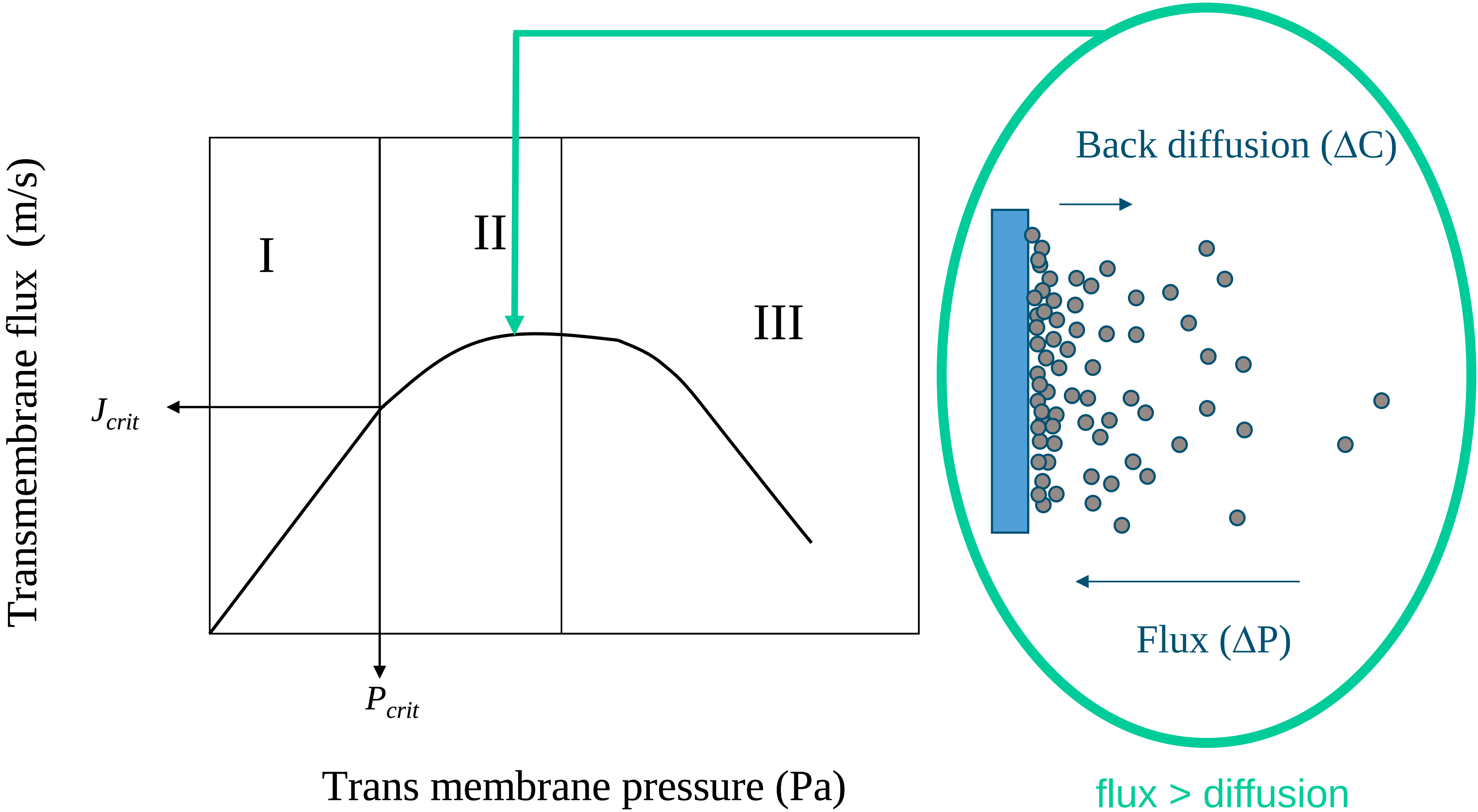


# Critical flux: starting point 'particles'



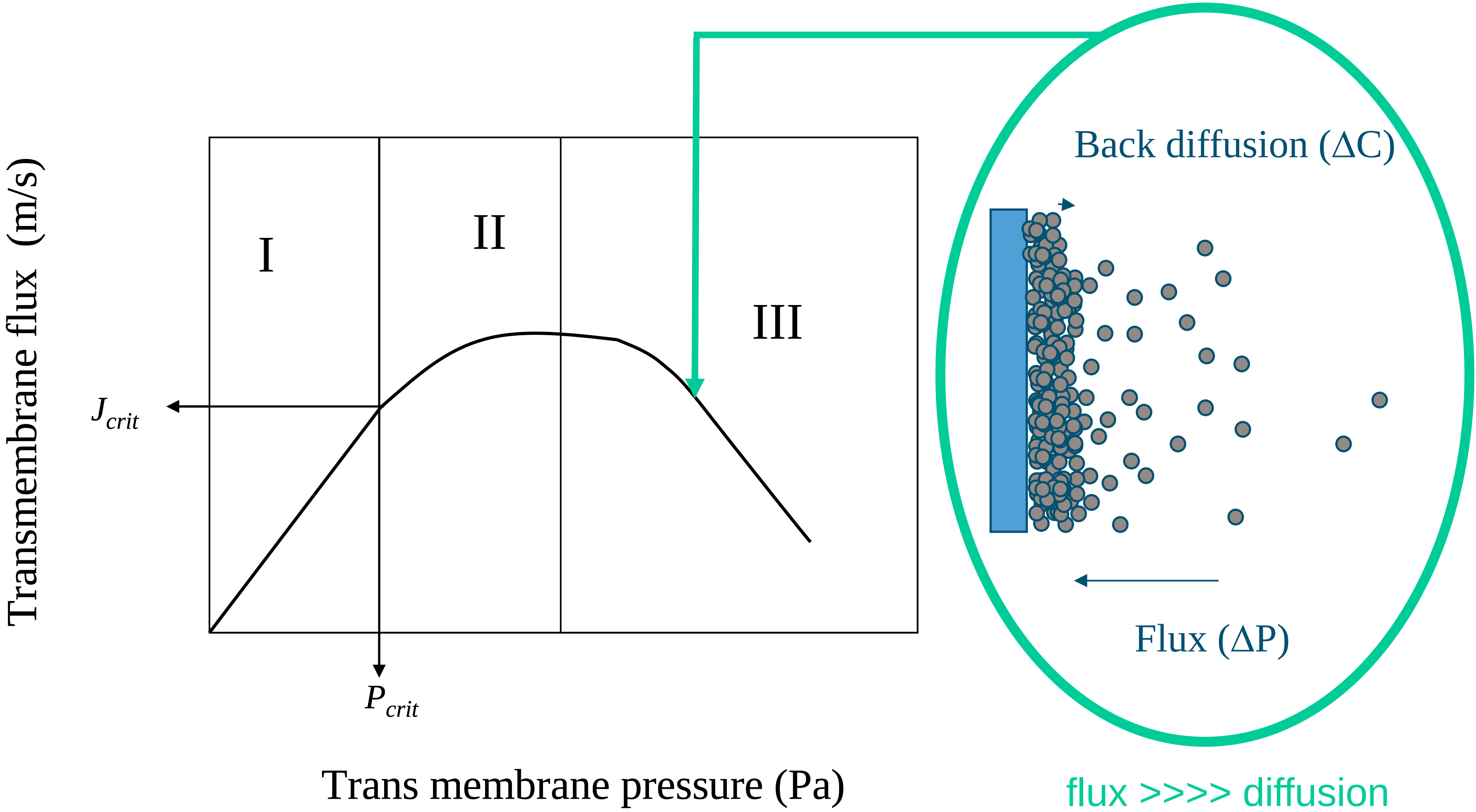

Delicate balance: flux ~ diffusion

# Critical flux: starting point 'particles'




flux > diffusion

# Critical flux: starting point 'particles'




flux >>>> diffusion

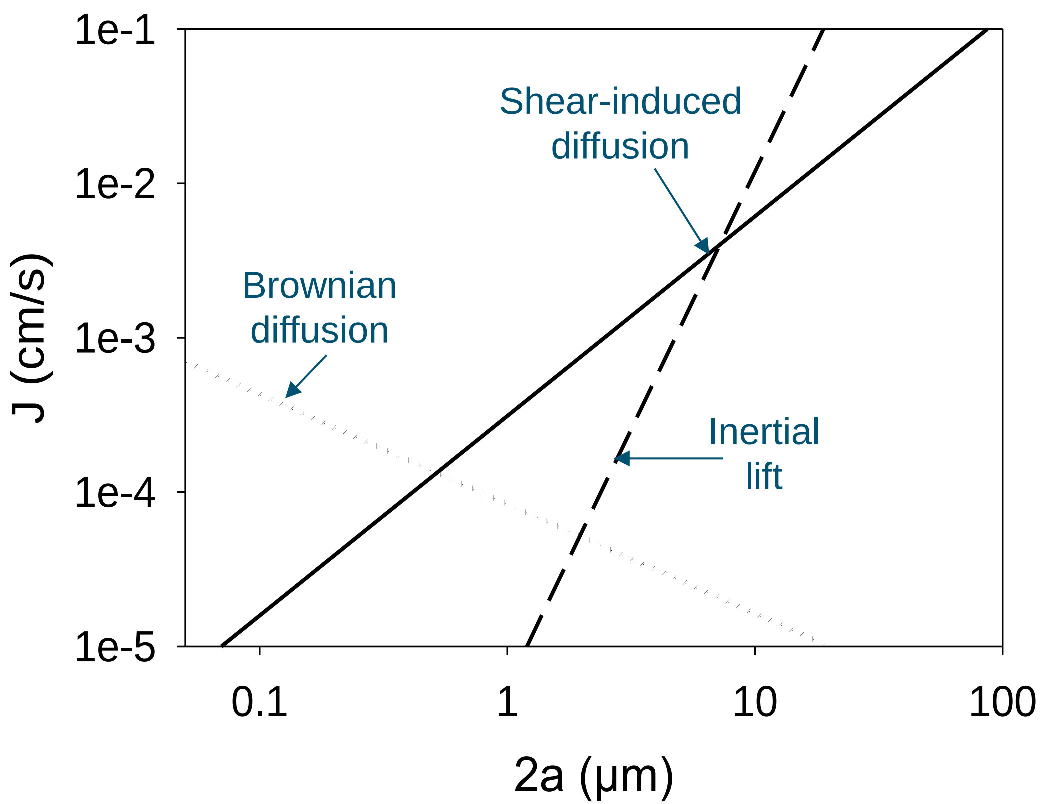
# Rationale for particles

Balance flux and back transport mechanism!

Particles of different sizes in milk

- Shear induced diffusion

UTP concept makes use of this, but can we do better?




*R.H. Davis, Separation and Purification Methods, 21, 1992*

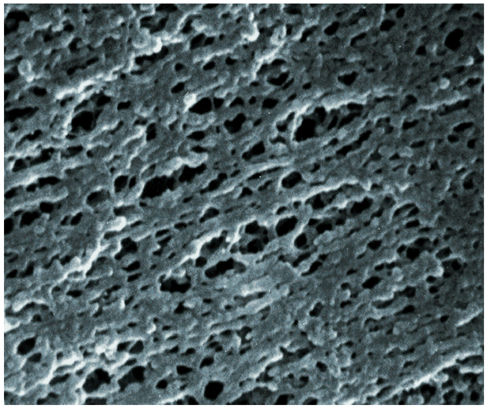
# Intermezzo: issues membrane design

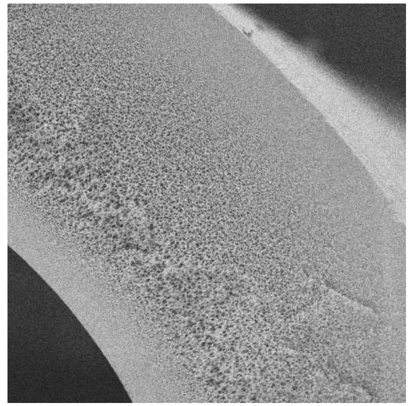
- Pore size distributions!
  - Local fluxes differ
  - Length of module
  - How to design a process?

➢ Membranes with uniform pores

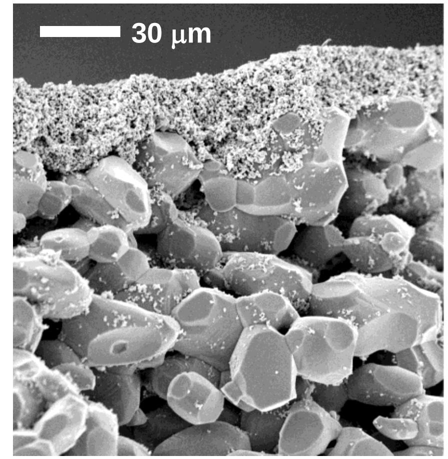
- Prerequisite: Surface properties
  - Adhesion/binding to the surface

# Membrane design: Pore size distribution

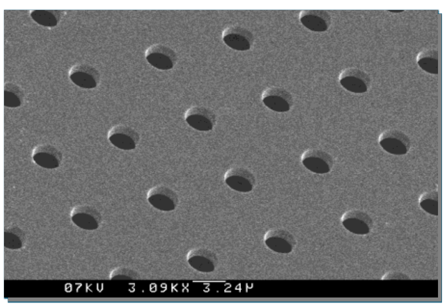




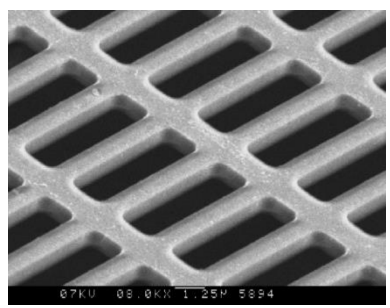
***Polymeric membrane (poly-sulfone)***



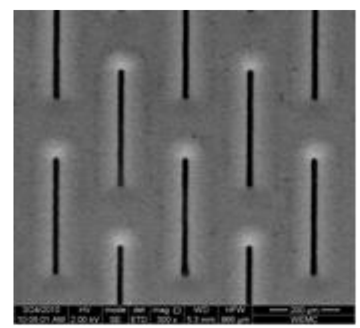

***Ceramic membrane***




***Micro sieve***
***Circular pores***

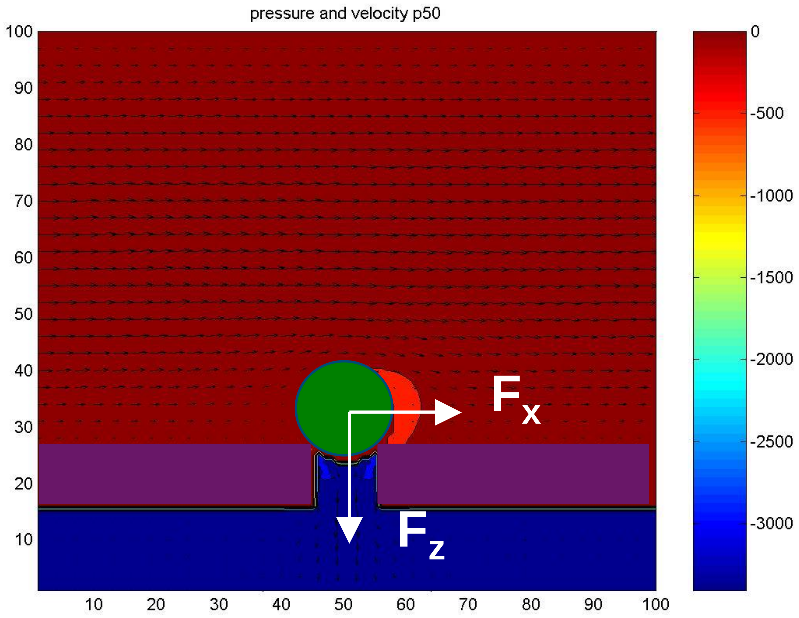


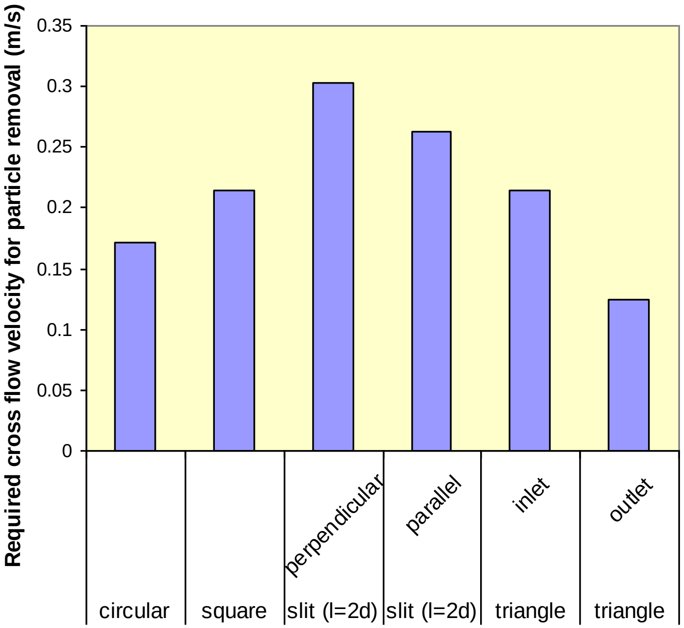

***Micro sieve***
***Slit shaped pores***



***Metal sieve***
***Slit shaped pores***

# Membrane design: Pore geometry




- Resulting force on a particle (drag, pressure, flow)

***TMP: 3.33 kPa; Particles 1.2 μm on 1 μm pore***

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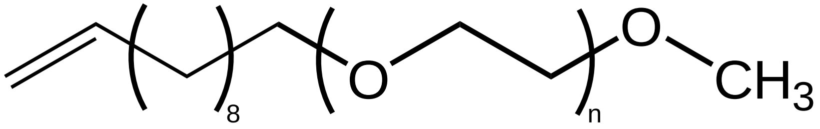
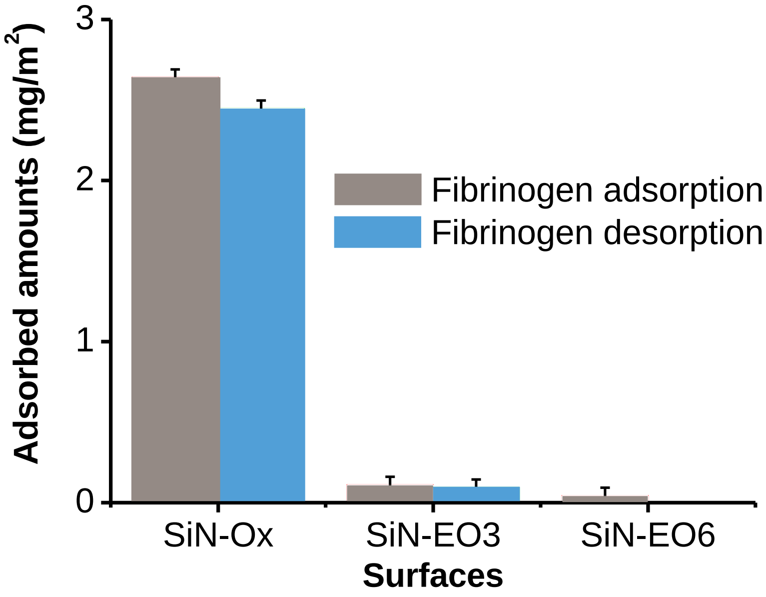
*G. Brans et al., Journal of membrane science, 2004*

# Membrane design: Surface properties

- Uniform pore size
  - High fluxes (but also high accumulation rate!)
  - Pore design & removal particles

- Surface properties
  - Mild modification
  - Length of molecule
  - Covalent bond






*Rosso et al. Langmuir 2008, 24, 4007-4012; Rosso PhD thesis, 2009, Wageningen*

# Back to the main issue

Prerequisites

- Pore size design
- Surface modification
- Uniform pores

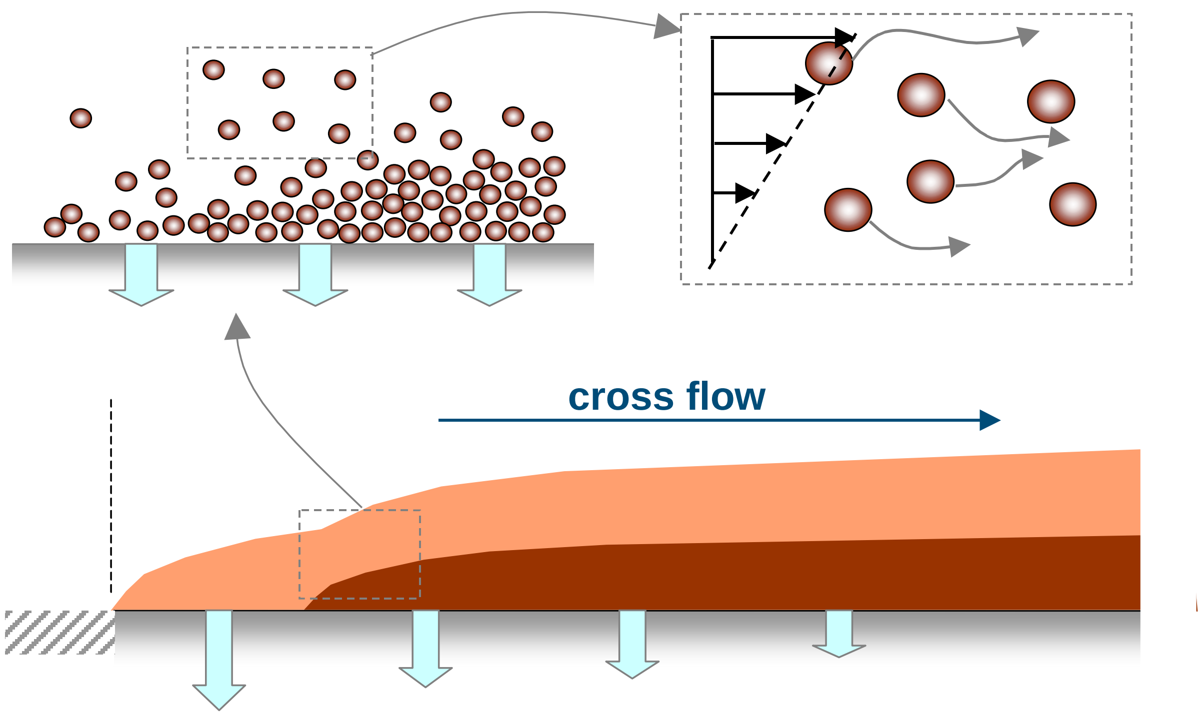
Core of the design

- ***Particle interaction in close proximity to the membrane***

Simulations as tools → experimental validation

# Back to particle behaviour

- Limiting process is concentration polarisation
- Simulation tools to predict behaviour (in complex feeds)

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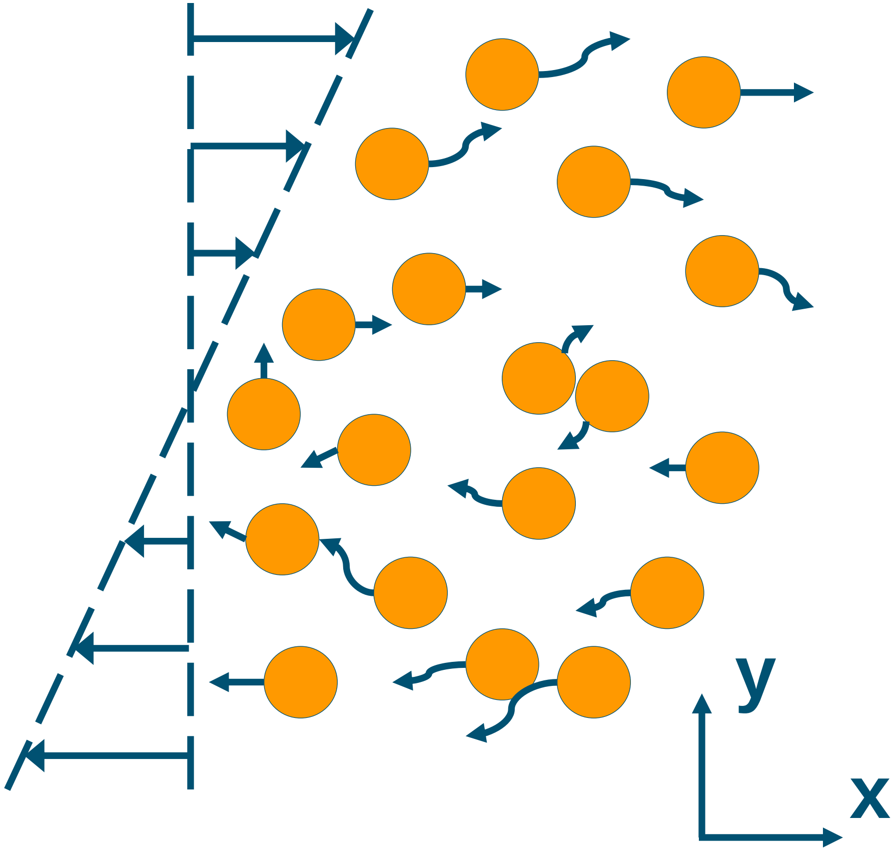
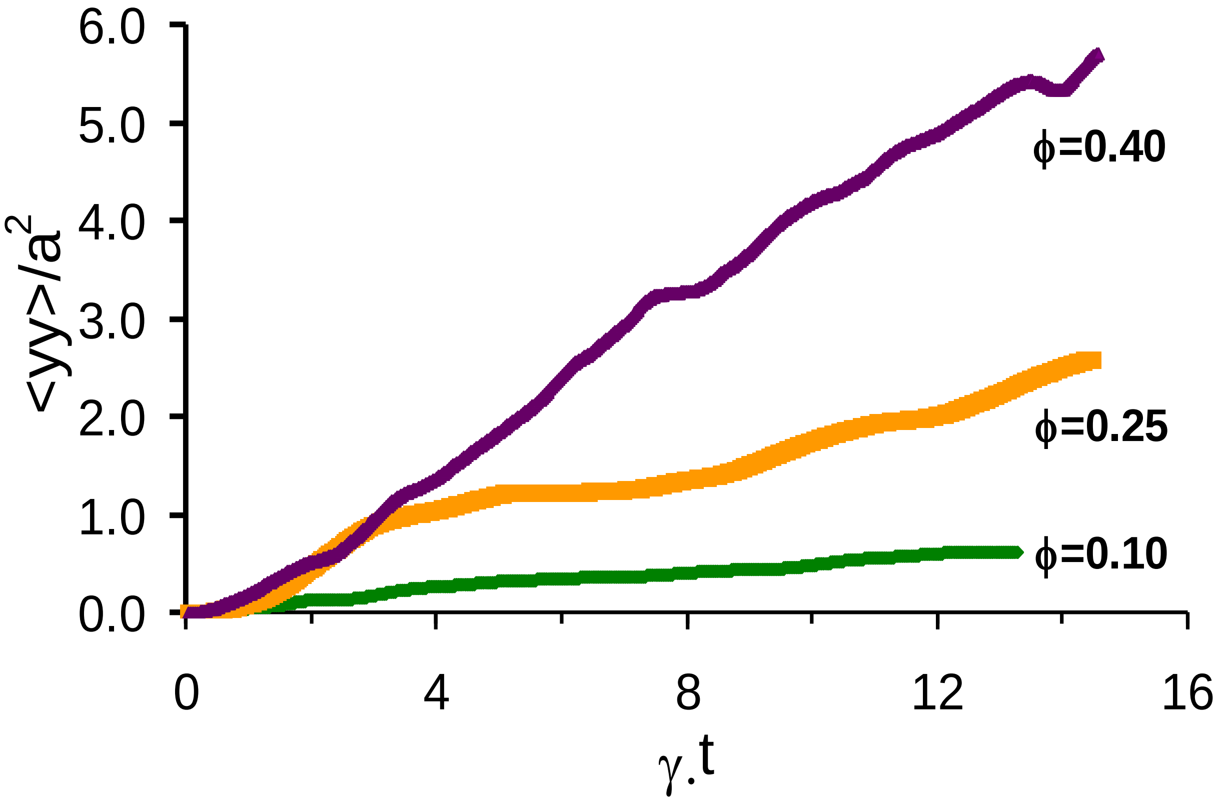
# Simulation of shear-induced self-diffusion

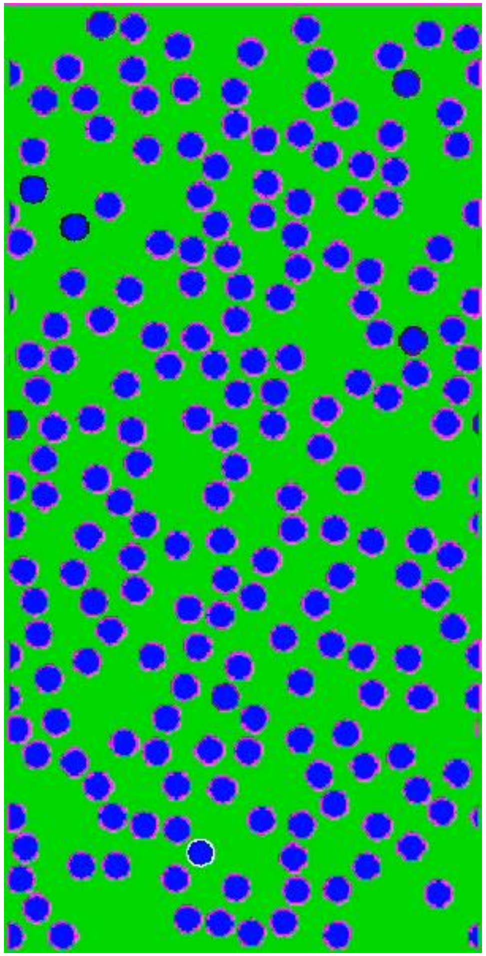
- Lattice Boltzmann method
- Suspension flow (Ladd, 1994)
- CFD-type approach
- Hydrodynamic interactions fully resolved
- Suspension particles considered as hard spheres
- 2-D simulations




*Ladd, Journal of Fluid Mechanics, (1994) 271 p. 285-309 and p. 311-339*

# Mean square displacement: mono-disperse



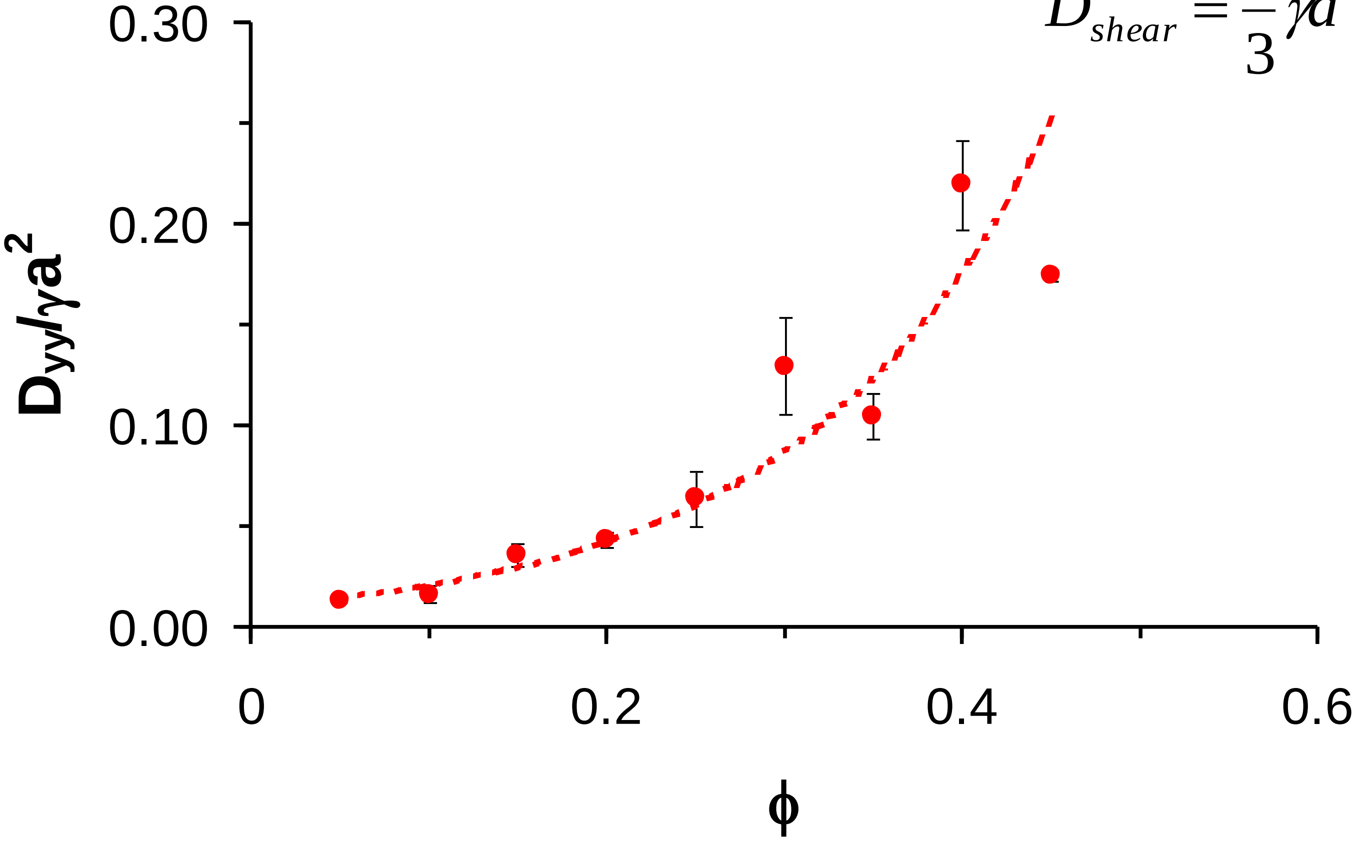





*Kromkamp et al, Journal of Fluid Mechanics (2005) 529, 253-278*

# Shear-induced diffusion: mono-disperse

$$D_{shear} = \frac{1}{3}\gamma a^2 \varphi^2 (1 + 0.5e^{8.8\varphi})$$

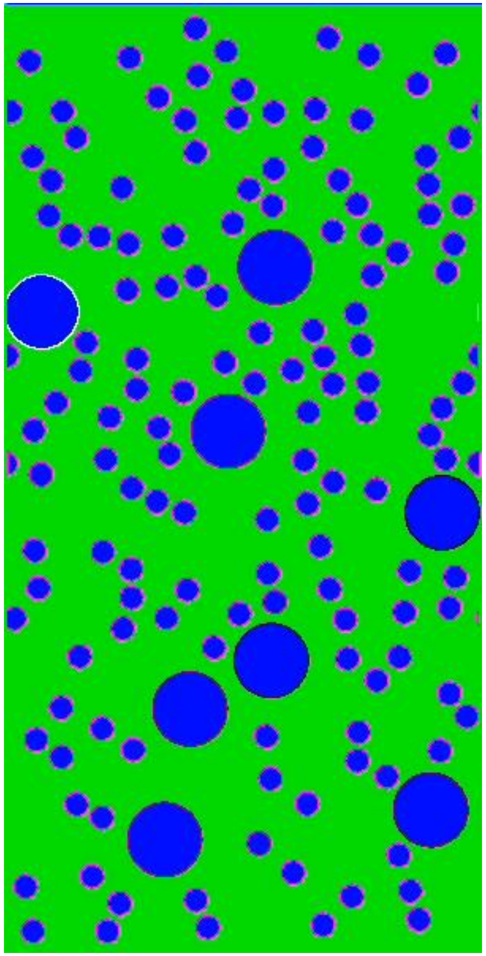


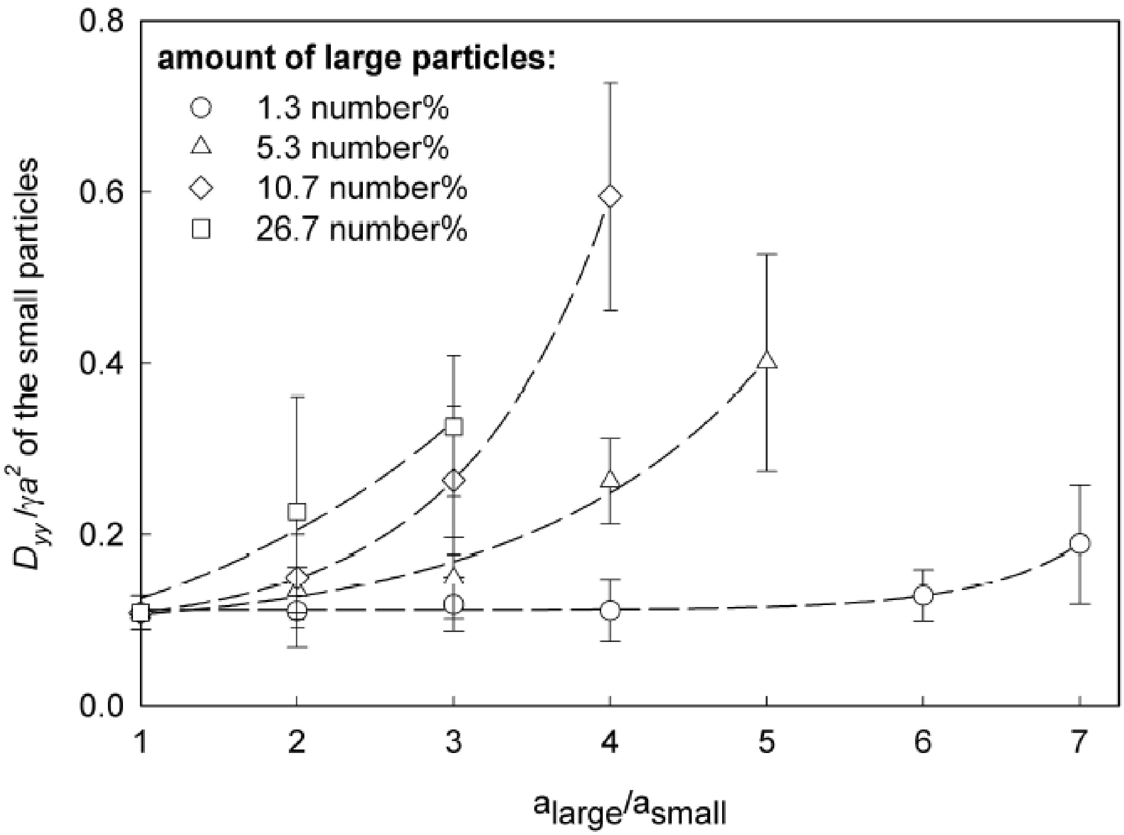

***Shear induced diffusion strong function of concentration***



*Kromkamp et al, Journal of Fluid Mechanics (2005) 529, 253-278*

# Shear-induced diffusion: bi-disperse






***Large particles dominate migration behaviour***

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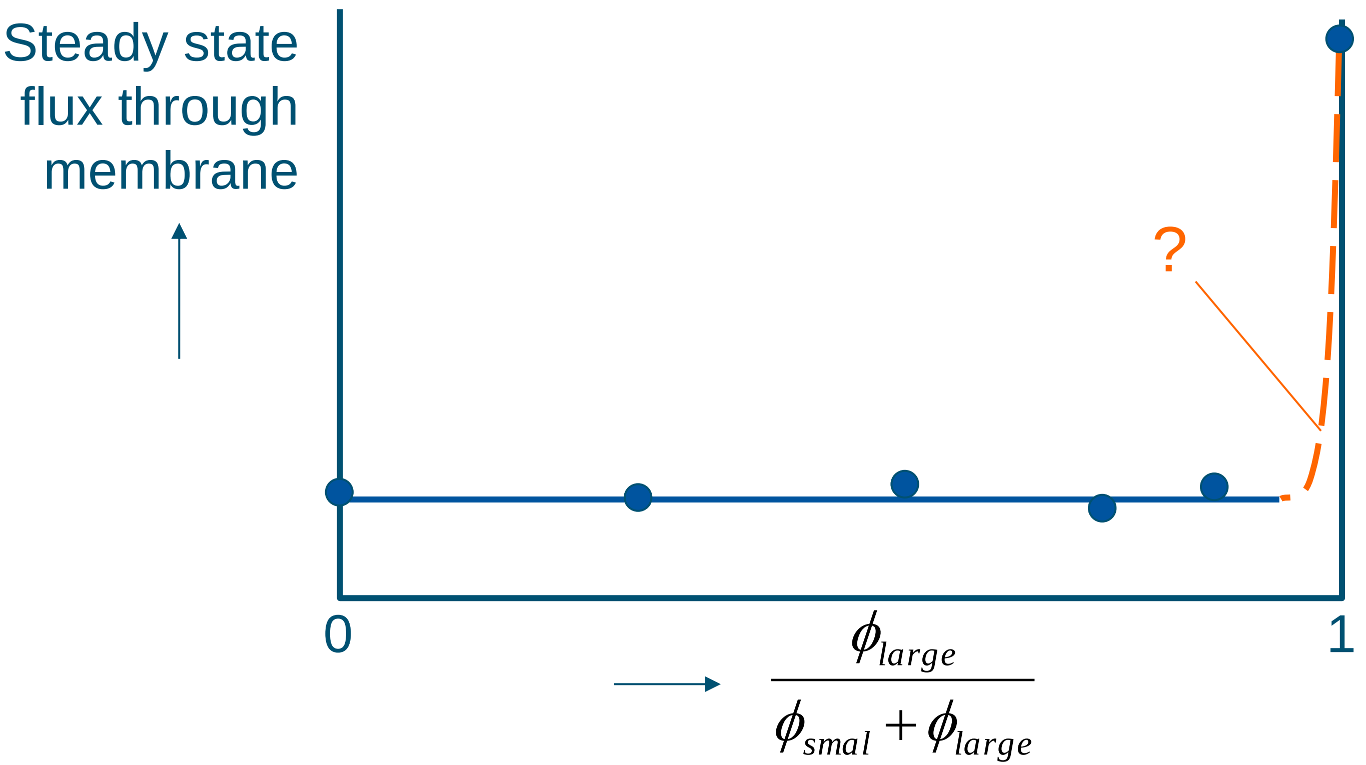
*Kromkamp et al, Journal of Fluid Mechanics (2005) 529, 253-278*

# Surprising flux behaviour




➢ ***Small particles dominate flux***

- Shear induced diffusion simulations point to reverse



*Kromkamp et al, Desalination (2002) 146, 63-68*

# Particle deposition

- So what is happening?

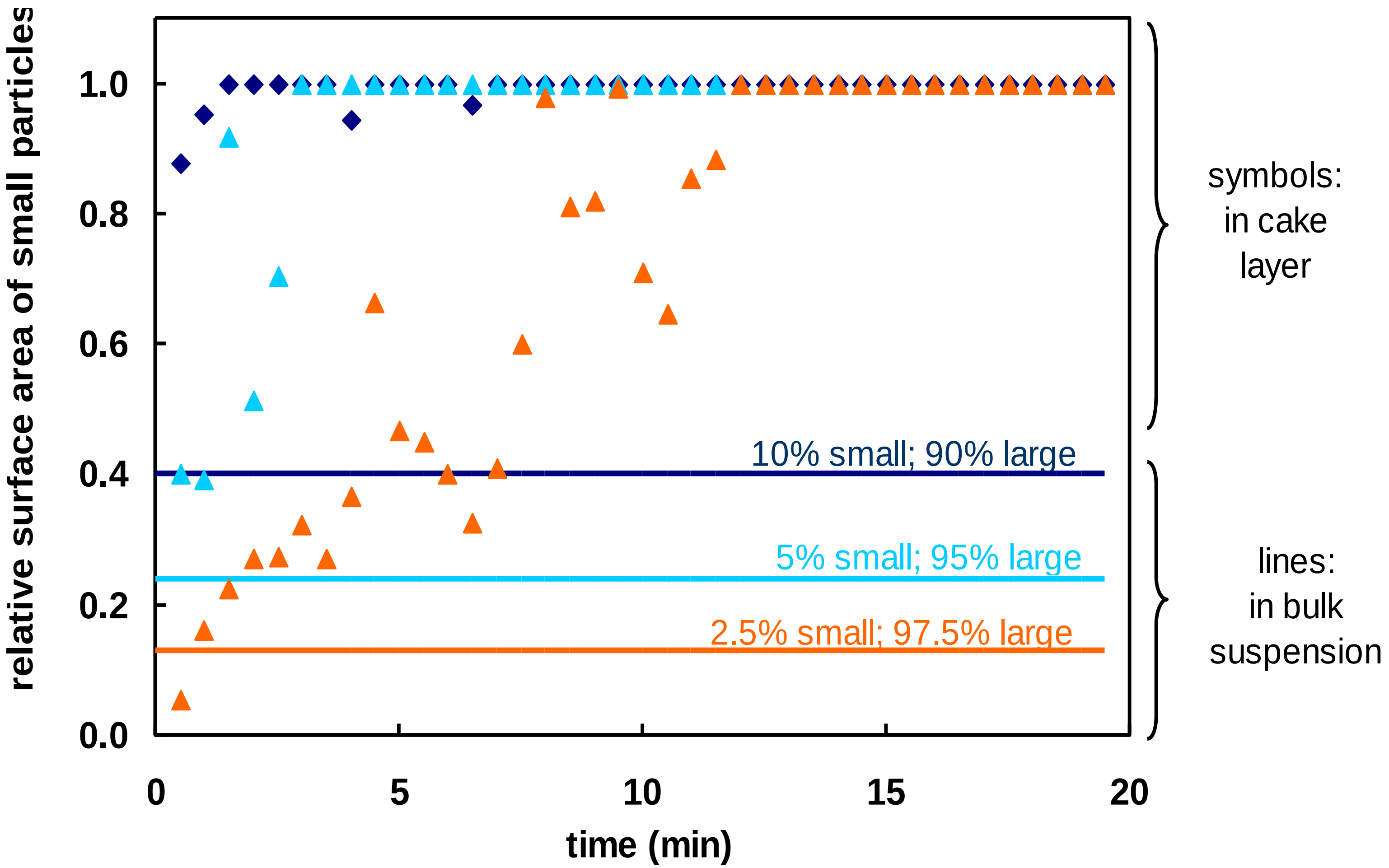
***Red large particles 9.7 µm (97.5%); Green small particles 1.6 µm (2.5%)***

***Membrane cut off 100 kDa, overlays of CSLM images***



*Kromkamp et al, 2006, Journal of Membrane Science*

# Relative amount small particles in cake




***Large particles 9.7 µm;  Small particles 1.6 µm***

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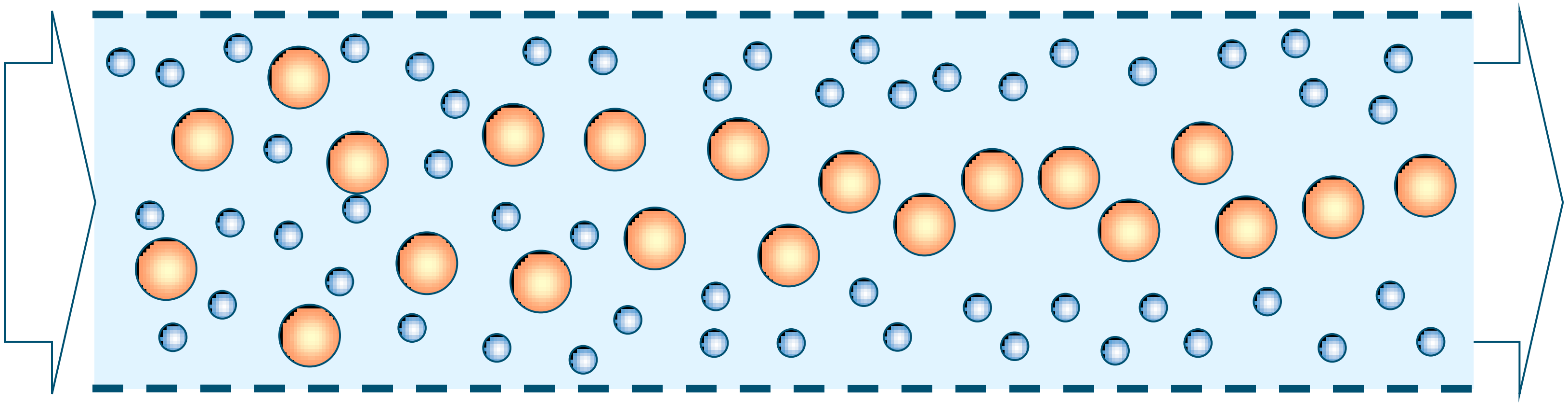
*Kromkamp et al, 2006, Journal of Membrane Science*

# Shear induced migration



- Hypothesis: Shear induced diffusion can lead to enrichment of small particles and depletion of large particles near membrane!

➢ ***Would confirm deposition & cake layer formation dominated by the small particles***

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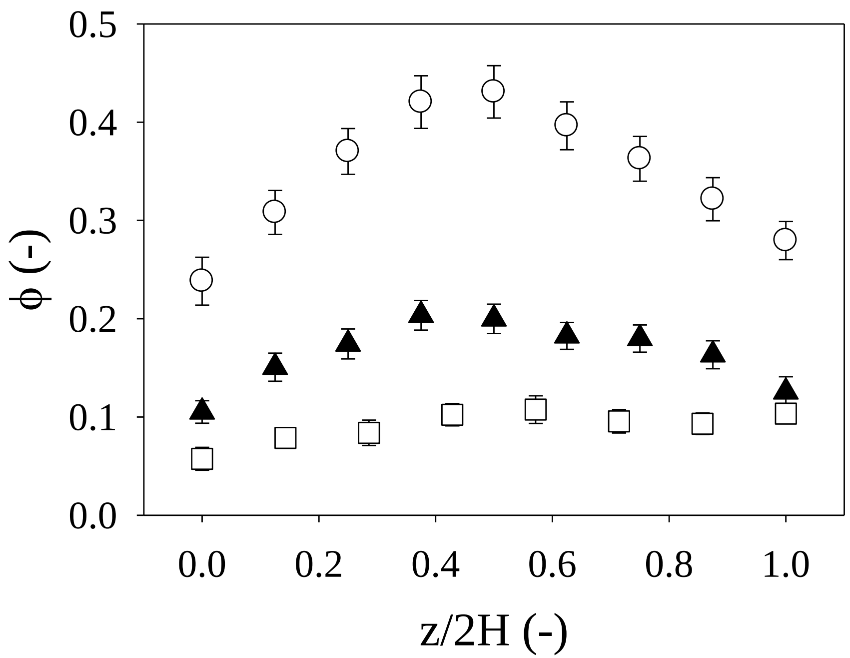
*Kromkamp et al, 2006, Journal of Membrane Science*

# CSLM: effect of concentration

- Quantify effects
  - CSLM in closed microchannel
  - Shear-induced diffusion only!

$$D_{shear} = \frac{1}{3}\gamma a^2 \varphi^2 (1 + 0.5e^{8.8\varphi})$$




***Concentration 2.65 µm particles in channel of 100 µm :***

***$\varphi_{tot}$ is 0.38 (○), 0.19 (▲), 0.09(□)***

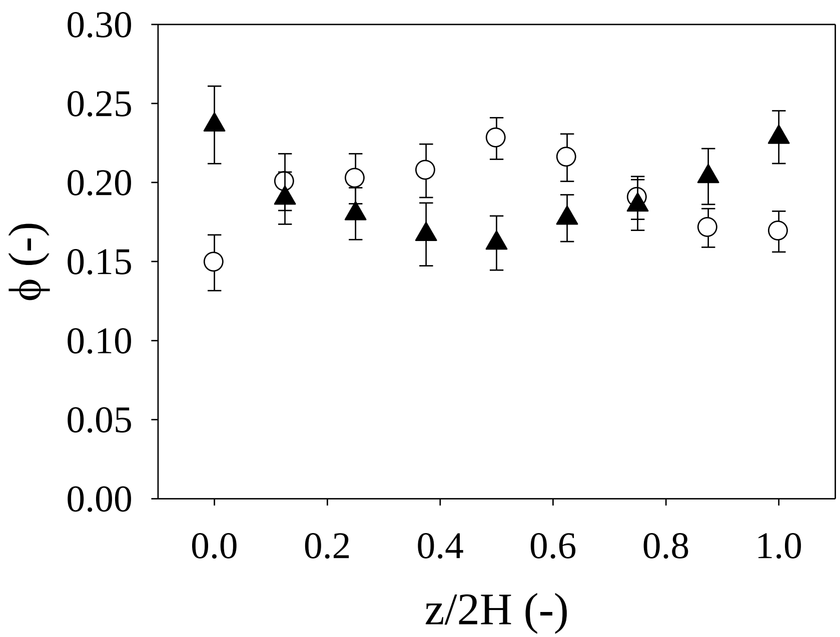
***v is 20.8 µm/s***

*van Dinther et al, Microfluidics and Nanofluidics 2013, 15, 451-465.*

# CSLM: effect of particle size

- Quantify effects
  - CSLM in closed microchannel
  - Shear-induced diffusion only!

$$D_{shear} = \frac{1}{3}\dot{\gamma} a^2 \varphi^2 (1 + 0.5 e^{8.8\varphi})$$




***Concentration large & small particles in channel of 100 μm :***

***$\varphi_{tot}$ is 0.38***
***(○) particles 1.53 μm***
***(▲) particles 2.65 μm***
***v is 20.8 μm/s***
***$\varphi_{small}/\varphi_{total}$ is 0.5***

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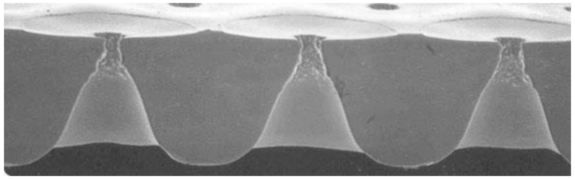
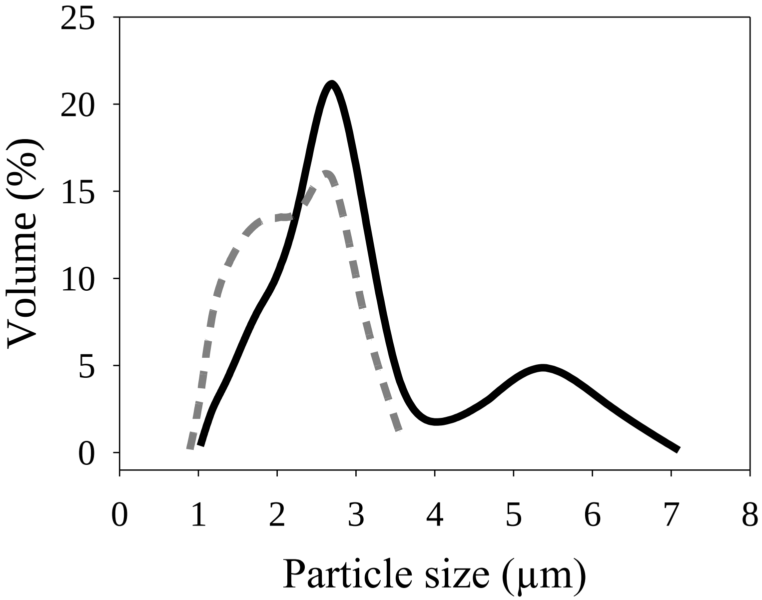
*van Dinther et al, Microfluidics and Nanofluidics 2013, 15, 451-465.*

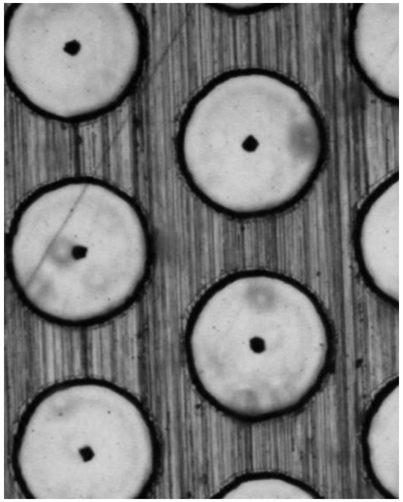
# Membrane fractionation (high concentrations)

- Quantify effect on emulsions
  - 20 *μm* pore size, metal sieve
  - channel height 200 micron






***Emulsions:***
***Small 1.35 µm,***
***Large 2.66 µm***
***$\varphi_{tot}$ is 0.27***
***$\varphi_S / \varphi_{tot}$ is 0.50***
***v is 0.59 m/s***
***Flux: 200-2200 L/m$^2$•h***



***Membrane pore >> droplet, no accumulation***

***Balance particle transfer to membrane and back diffusion***

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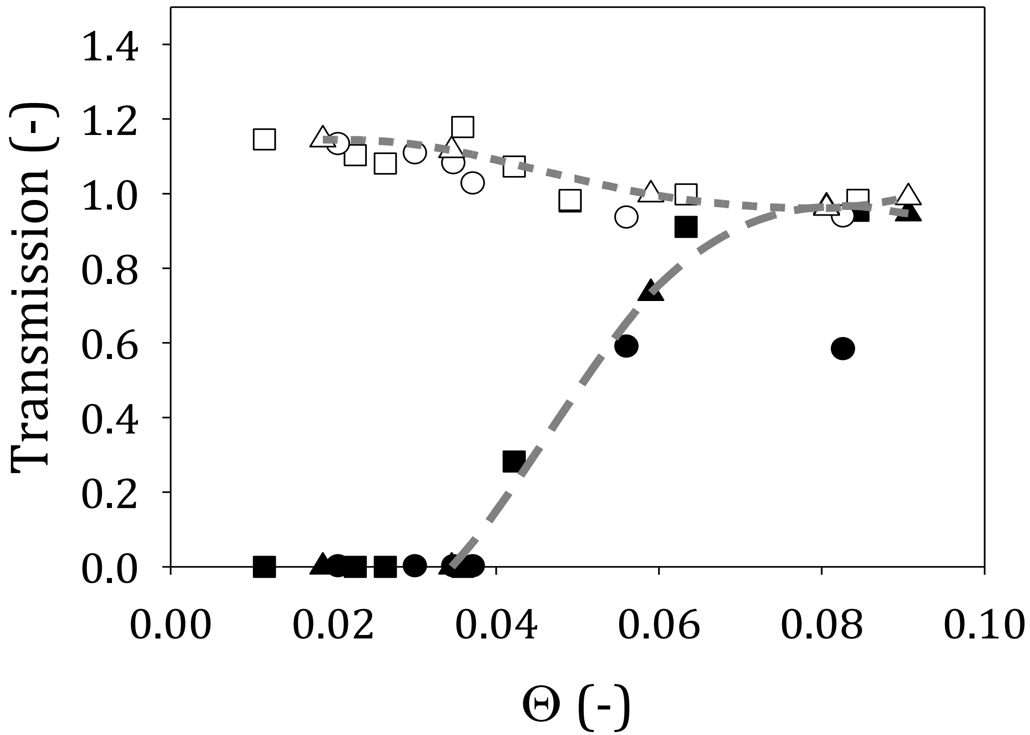
*van Dinther et al, Innovative Food Science and Emerging Technologies 18 (2013) 177–182*

# A closer look at fractionation




***Volume fraction 0.37***
***Small 1.35 μm,***
***Large 2.66 μm***
***$\varphi_S / \varphi_{tot}$ is 0.50***

- Depending on cross-flow versus trans-membrane velocity
  - ***Only small particles in permeate***
  - ***Sometimes at higher concentration than in the feed!***

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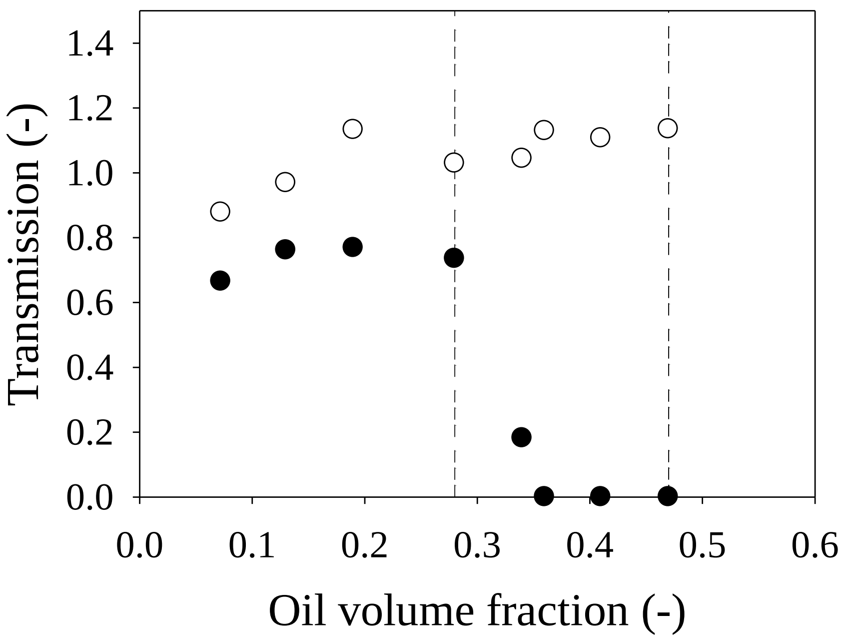
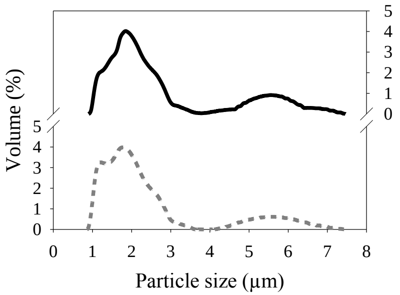
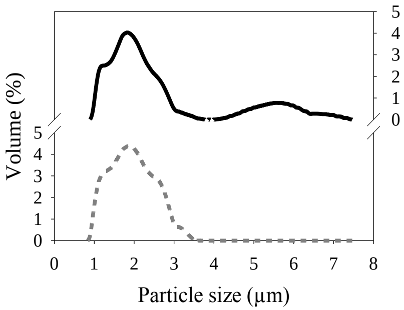
*van Dinther et al, Innovative Food Science and Emerging Technologies 18 (2013) 177–182*

# Effect of concentration




***Volume fraction 0.28***
***Feed***
***Permeate***




***Volume fraction 0.47***
***Feed***
***Permeate***

***Higher concentration works better, till certain limit!***
***Shear induced diffusion is more pronounced***

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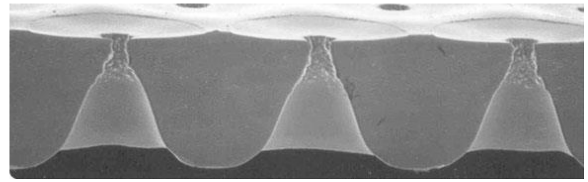
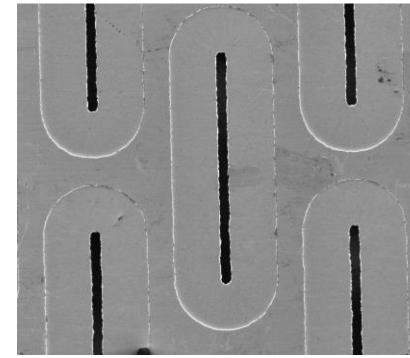
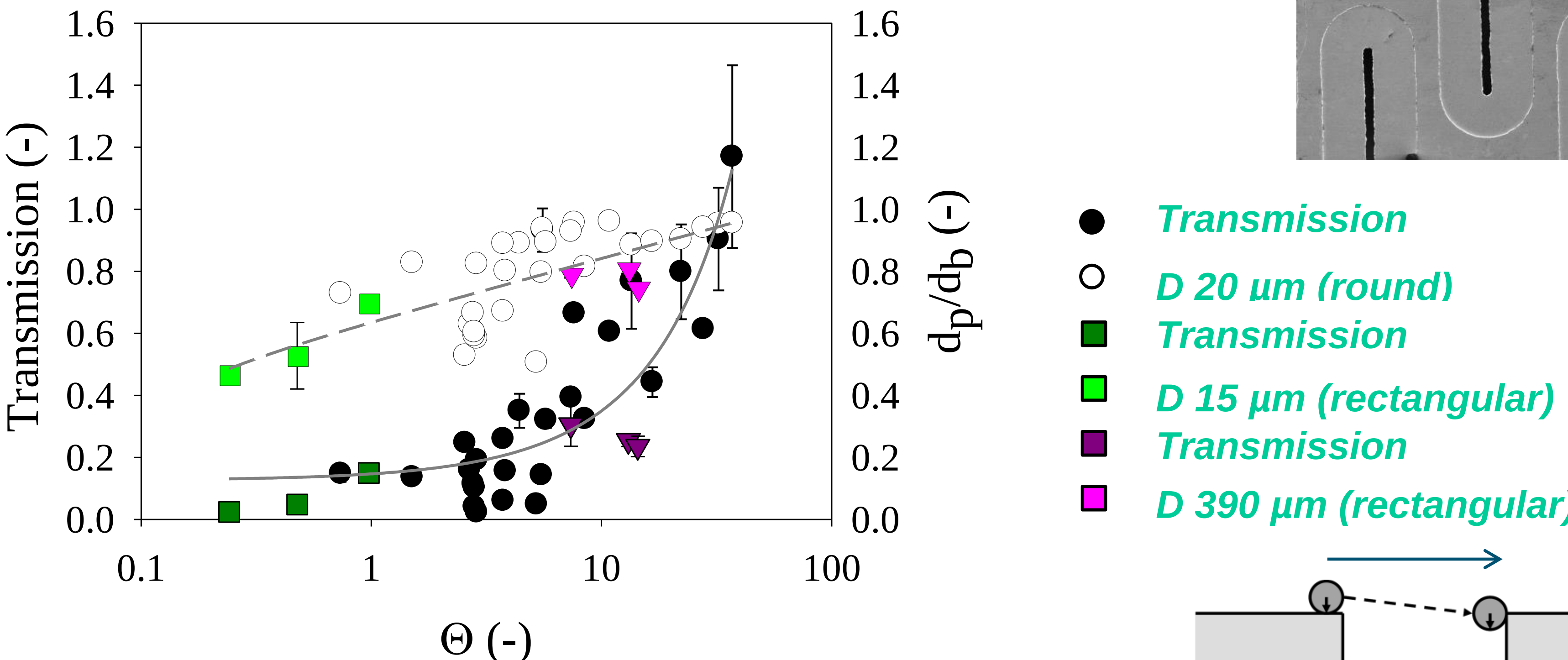
*van Dinther et al, Innovative Food Science and Emerging Technologies 18 (2013) 177–182*

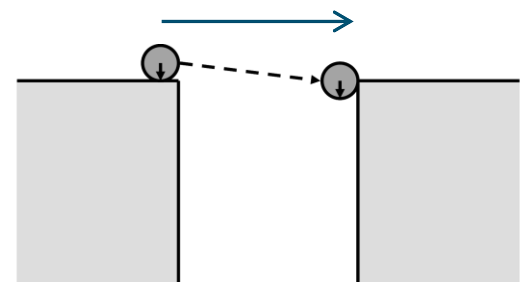
# Membrane fractionation (low concentration)

- Various metal sieves







➢ ***Skimming: Only small particles in permeate***

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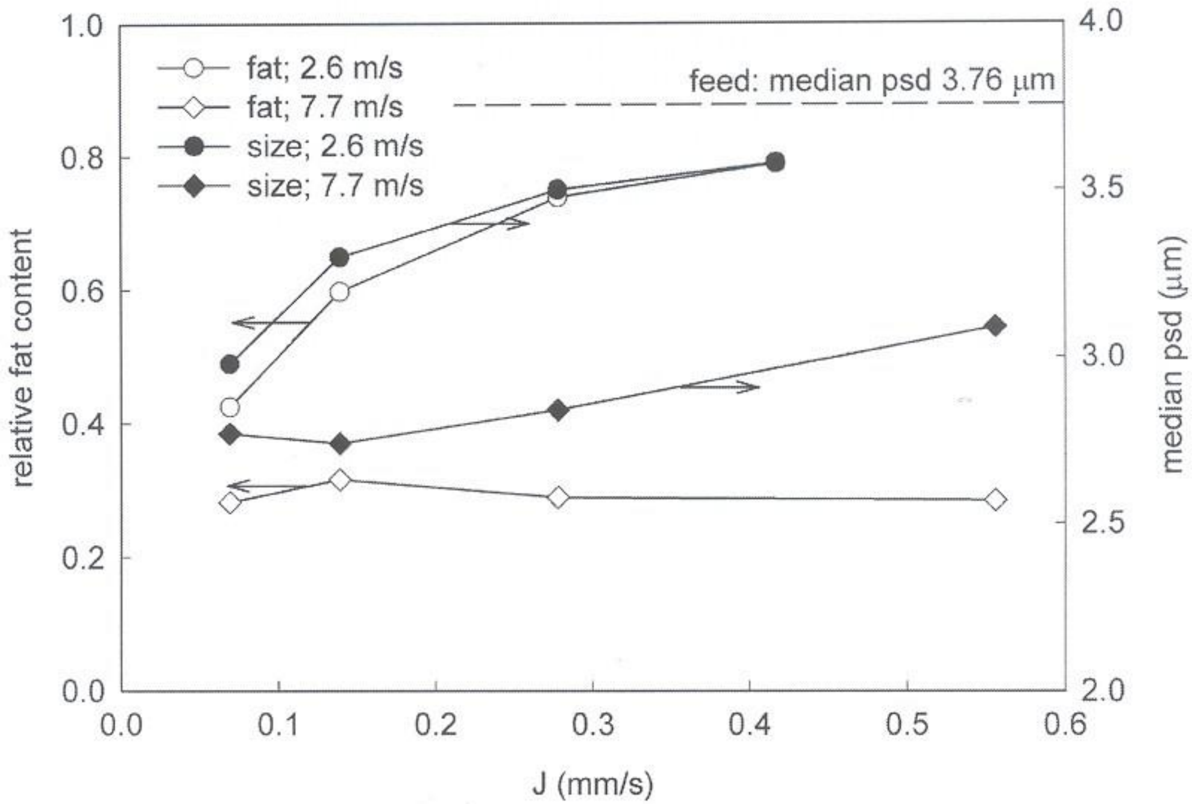
*Dinther, et al. (2011) Journal of Membrane Science 371 (1-2). - p. 20 - 27.*

# Membrane fractionation of milk fat




- Pore size 5 μm
- Deposit free operation
- CFV and flux determine composition permeate
- Pore size not so relevant

➢ ***Option: pore size > largest particle***

*Kromkamp et al, Journal of Membrane Science, 2006*

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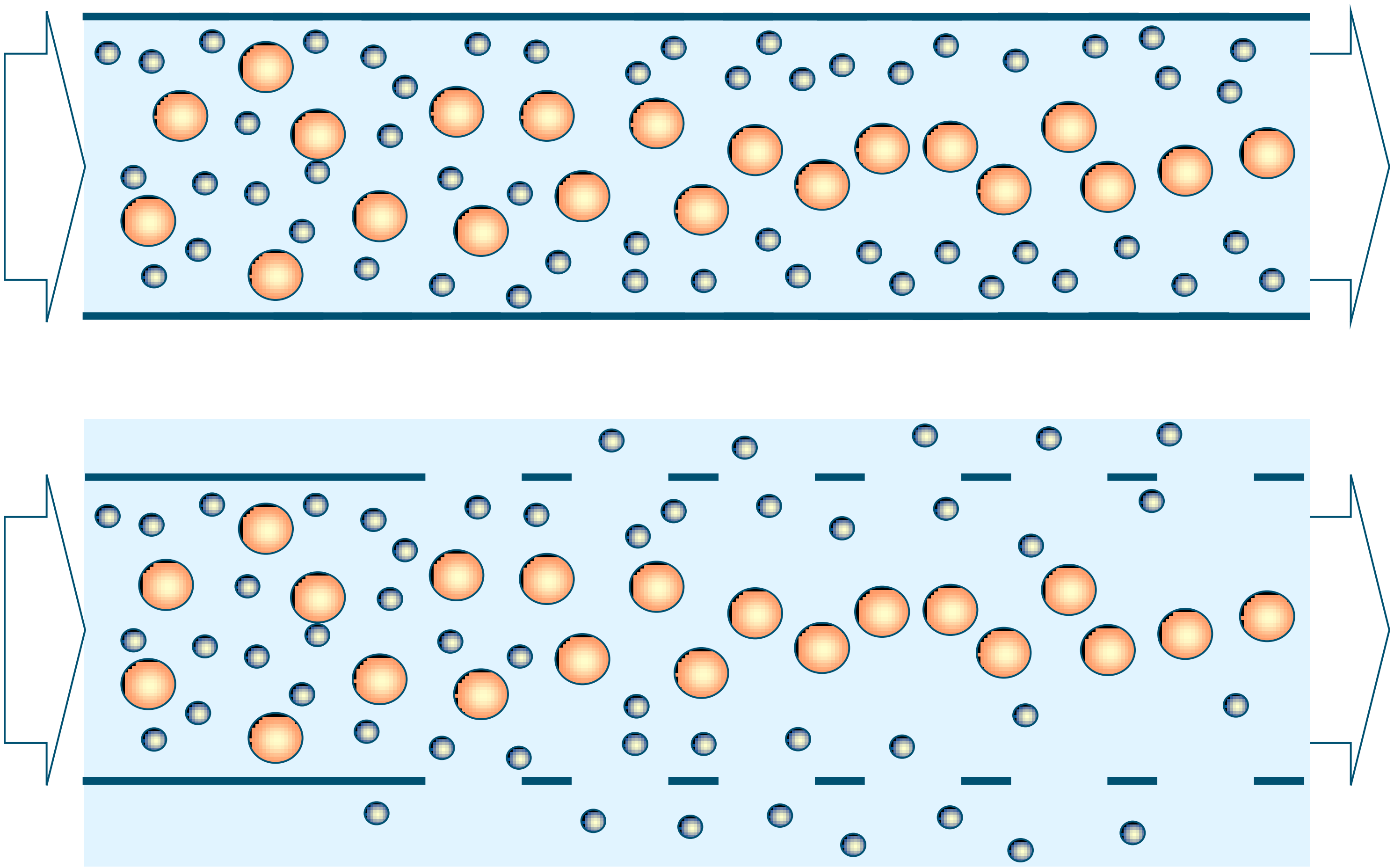
# Fractionation with large(r) pores

- ***Opportunities: 'only' process control is needed!***



# Conclusions / implication process design

- Detailed investigations into particle behaviour:
  - Diffusion highly concentration dependent
  - Large particles dominate diffusive behaviour
  - Smallest particles dominate flux behaviour (depending on pore size)
  - Large pores: Selectivity dependents on process conditions

- Processes could be controlled through cross flow and pressure
  - Choice of pore size may be not too important….
  - Fouling could be better controlled
  - High fluxes are possible during continuous operation

# What is needed to get this to work?

- Entrance length for sufficient migration (1 mm - 50 cm)
- Large uniform pores >> particles
- Process conditions chosen based on
  - Diluted systems → fluid skimming
  - Concentrated systems → shear induced diffusion

- High fluxes and no fouling
- Yeast, emulsions, algae, gel beads: Principle works!
- Fractionation of particles very close in sizes possible

# Thank you for your attention

Pore size may not always be that relevant.....but process conditions are

Micrometer scale insights are needed for radically new process designs

Keep looking for unexpected options!

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***More info: karin.schroen@wur.nl***