Composite Technology MSE 440

Background in process modelling

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Objectives

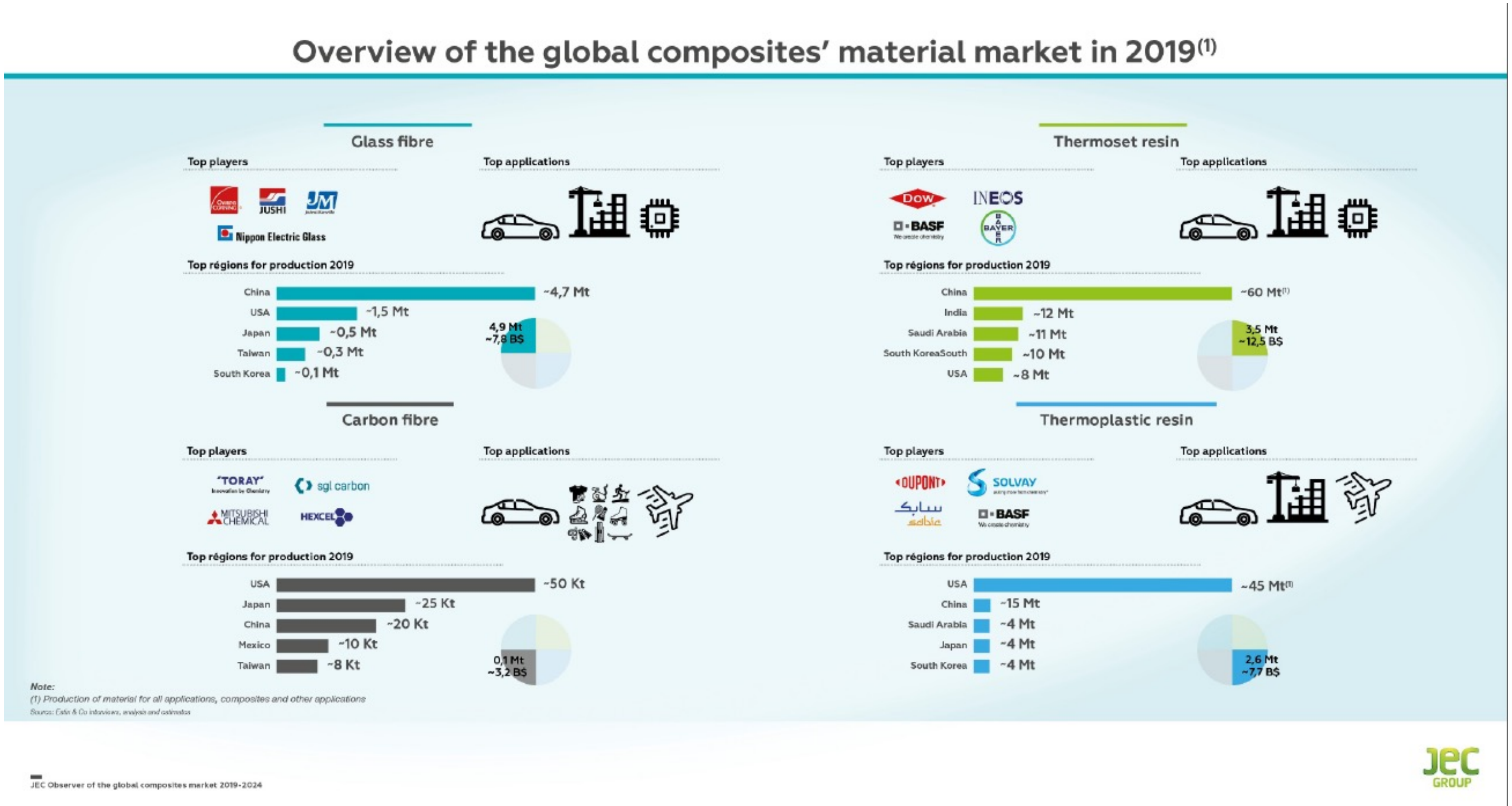
- Justify the choice of a given process based on the materials selection, part complexity and production volume.

- Know the basic flow equations and evaluate the time to process a simple part by Liquid Composite Moulding.

- Explain the parameters governing the process kinetics and propose strategies to optimize part processing

Composite materials processing

Last week, you saw the main constituents (fibers, polymers) and the main processing methods.



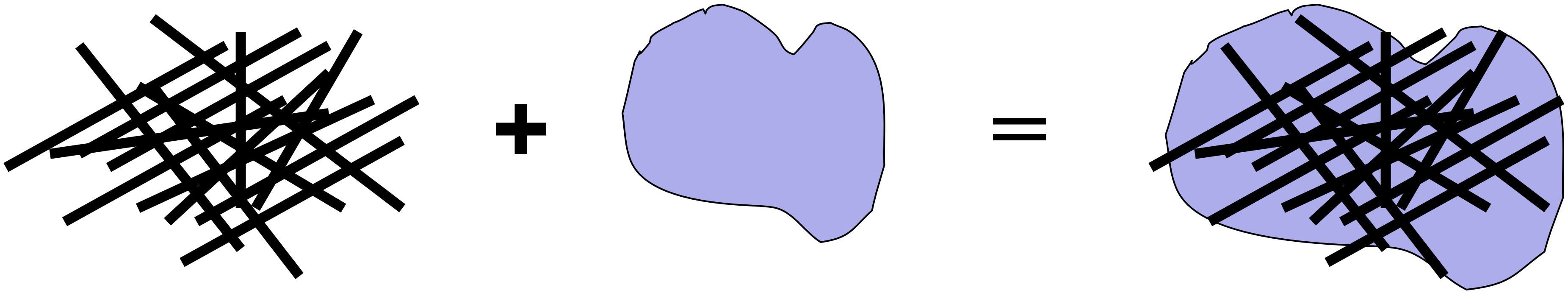
Composite processing

In general, the reinforcement is already under its definite form:
fiber, powder, potentially woven or assembled.
-> the matrix is the one that transforms during processing
-> Interface is also formed during processing.

Processing must ensure:

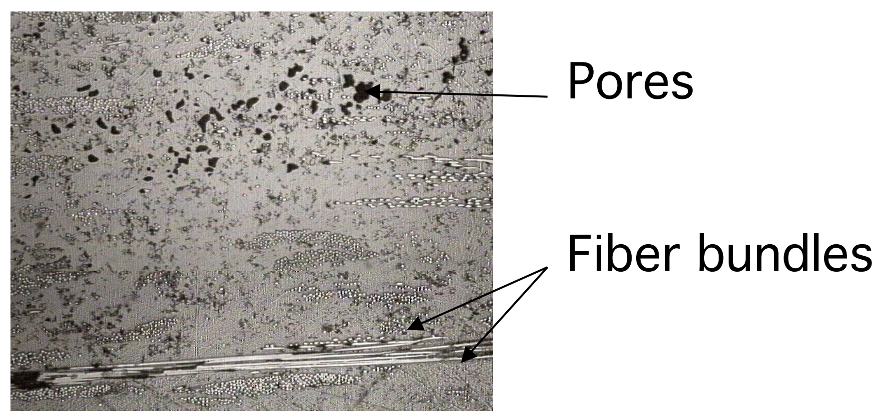
- to preserve the integrity of the reinforcement, its orientation, its architecture if needed.
- to provide a matrix that intimately bonds to the reinforcement, develops desired mechanical properties, and presents few defects.
- to be as cheap as possible, and as fast as possible to process for a given set of requirements.

Composite processing issues



Fundamentals:
- thermodynamics, interface chemistry
- multiphase flow in porous media
- heat and mass transfer (solidification, chemical reaction)
- mechanics of multiphase materials

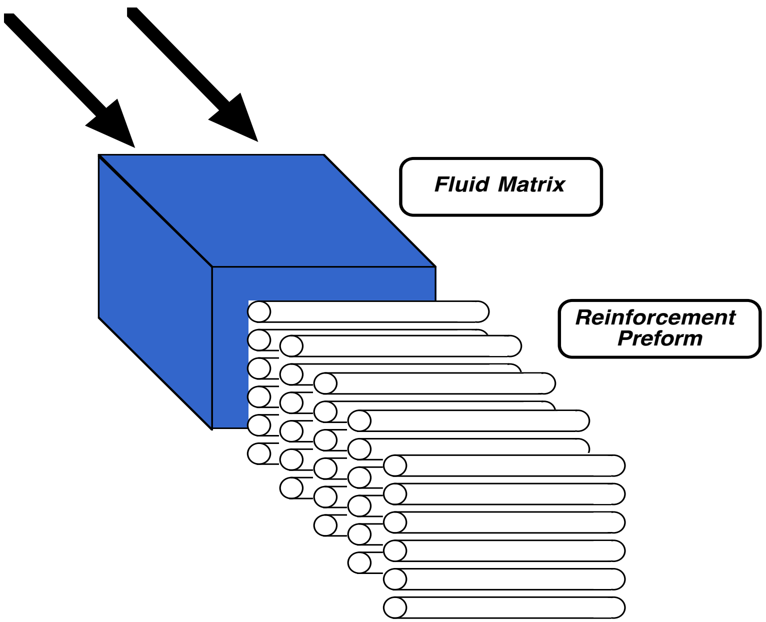
Materials quality implications:
- inhomogeneity
- porosity
- internal stresses
- lack of reproducibility



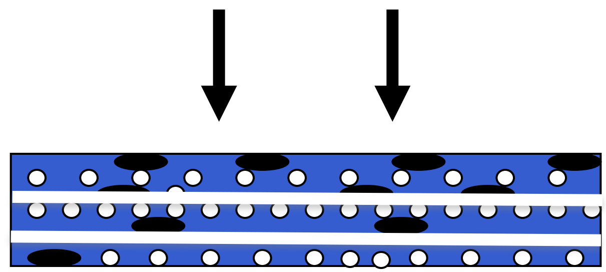
Modelling

In all composite processing techniques, the phenomena are flow of one or more fluids into a potentially compressible preform, with potential heat transfer.

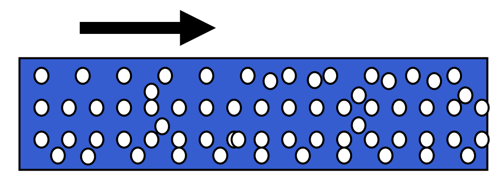
3 main cases, infiltration and consolidation (cf soil science) as well as flow of a suspension in the case of low aspect ratio materials:



Infiltration

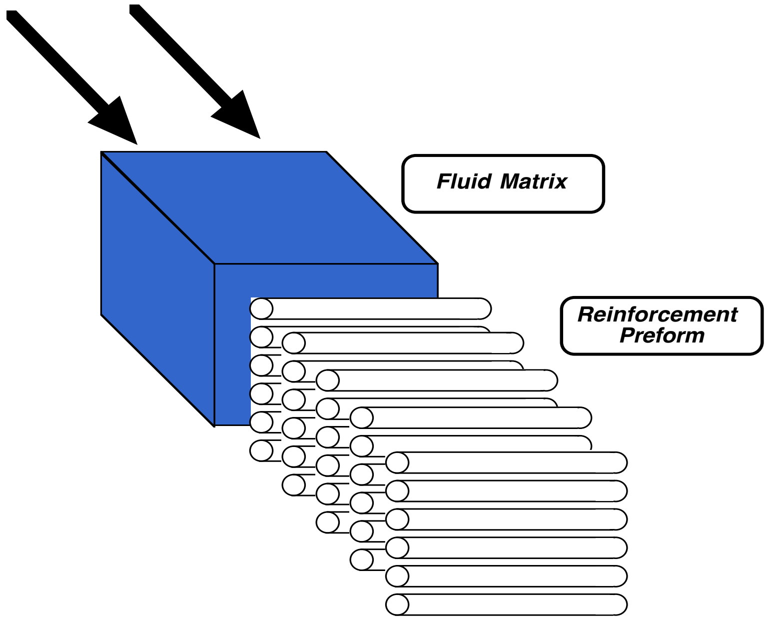


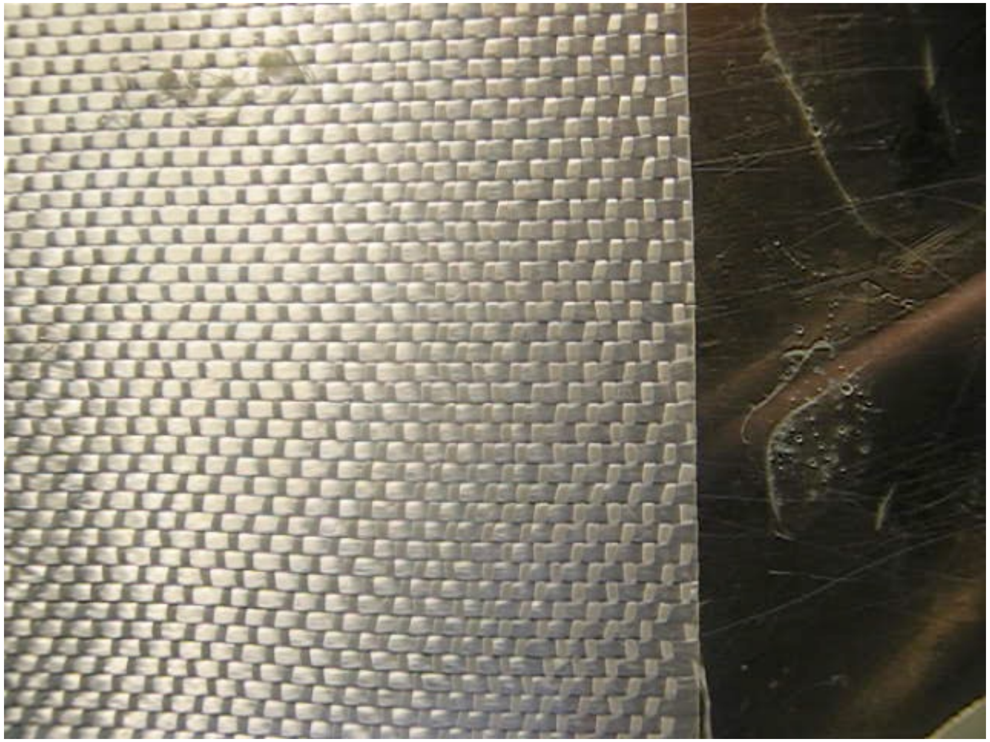
Consolidation



Suspension flow

Example of liquid composite molding





Modelling- Physical phenomena

Equations describing the physical phenomena:
- Fluid flow into a porous medium or flow of a suspension
- Mass conservation, possibly compressible fluid
- Heat transfer
- Phase change (solidification)
- mechanical forces

-Size of the reinforcement is in general small compared to that of the part -> continuum mechanics approach

-Definition of a representative volume element, small enough to contain averaged values, but large enough to contain a representation of the system (fiber and matrix, for example).

Modelling- constitutive equations

Constitutive equations of the parameters needed to solve the equations, which can also change with time, Temperature, Pressure, velocity:

-Resin: viscosity, cure kinetics, solidification

-Fibre bed: permeability, stress-strain behavior

- Resin/fibre interaction: thermodynamics, dynamic capillary effects

Boundary conditions : T mould, Applied pressure or flow rate, etc…

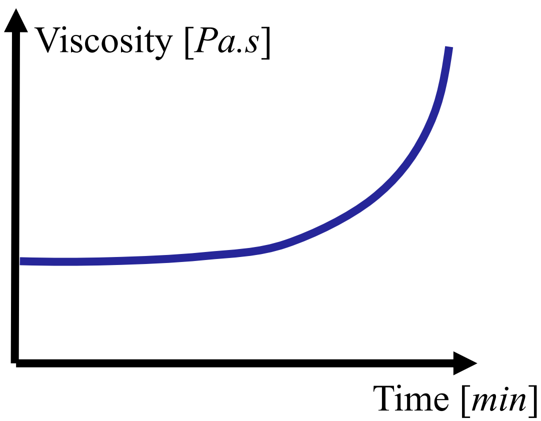
Experimental validation

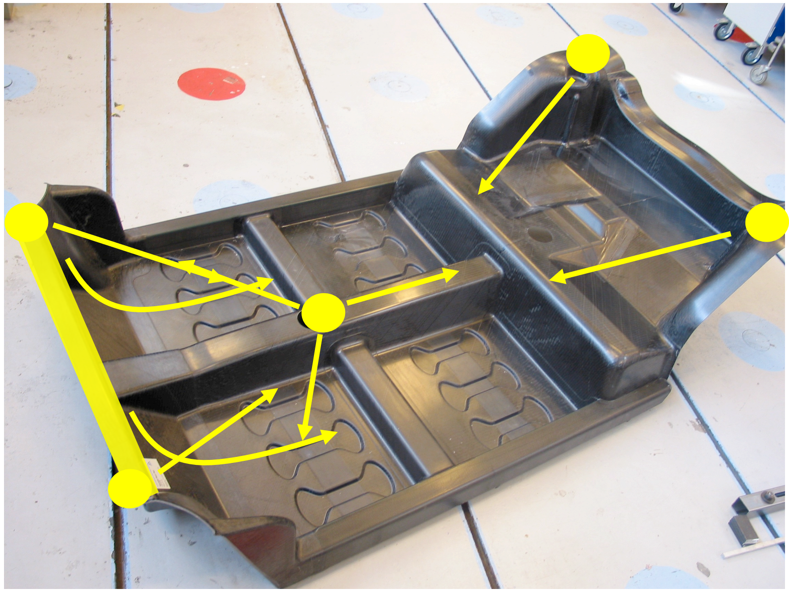
Goal of the models

Define processing windows for a given part production (polymer, fibres):

Role of Pressure, Temperature, geometric location of the resin inlets, method of resin delivery (pressured pot, pump..)

On the kinetics of resin progression (time to impregnate totally), on the final part quality.



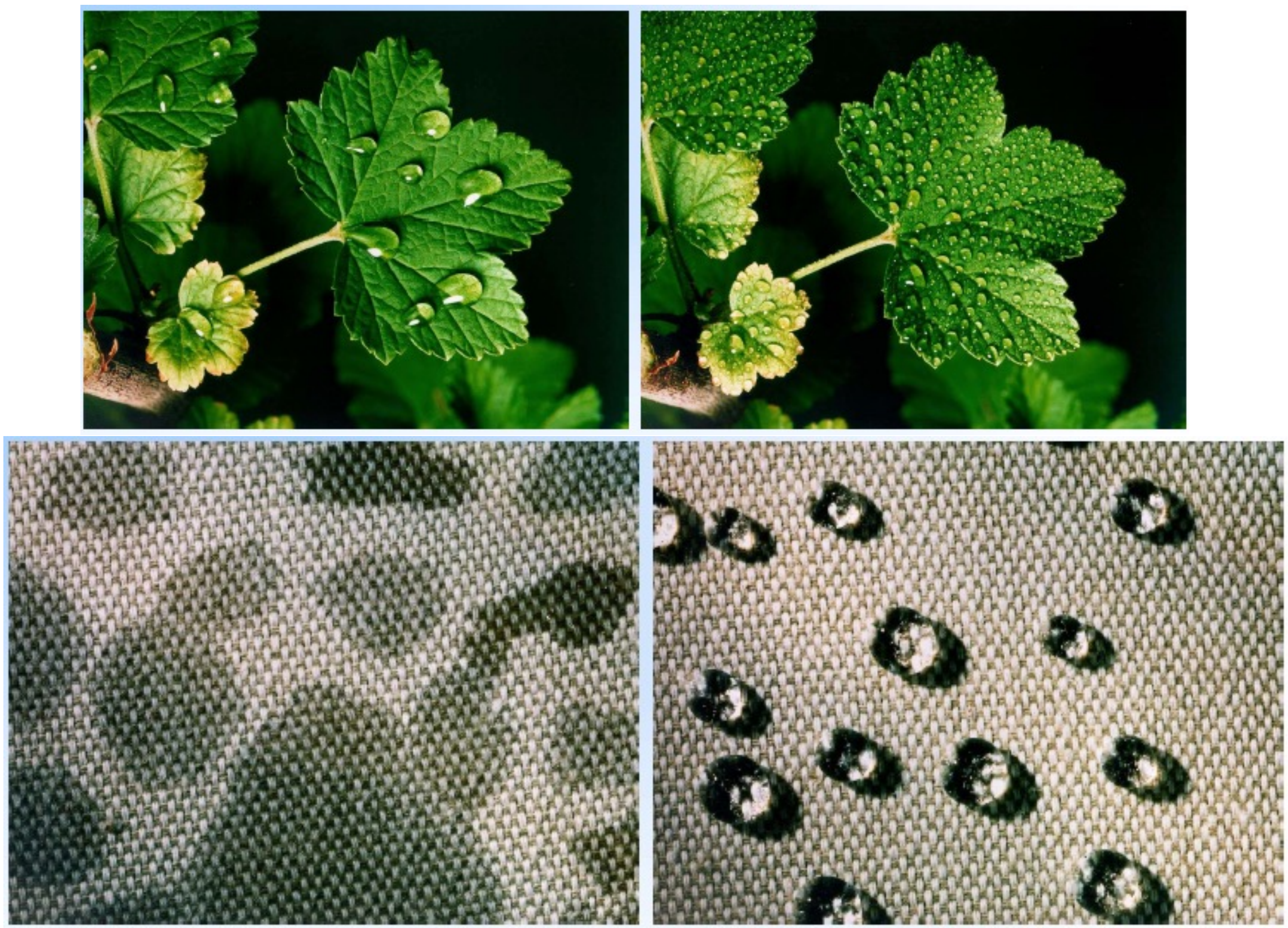


1 hour? 10 hours?
1 bar pressure? 10 bars?

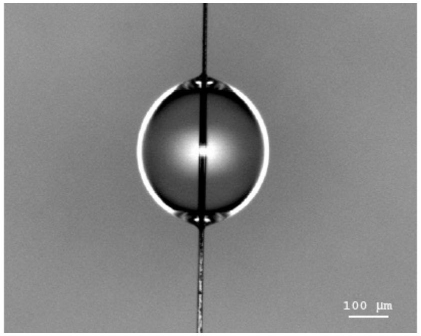
Flow modeling is necessary to ensure the injection is as fast as possible
- complete before viscosity build-up
- To achieve high production rate

Capillary phenomena

First question: does the resin spontaneously wet the fibers?

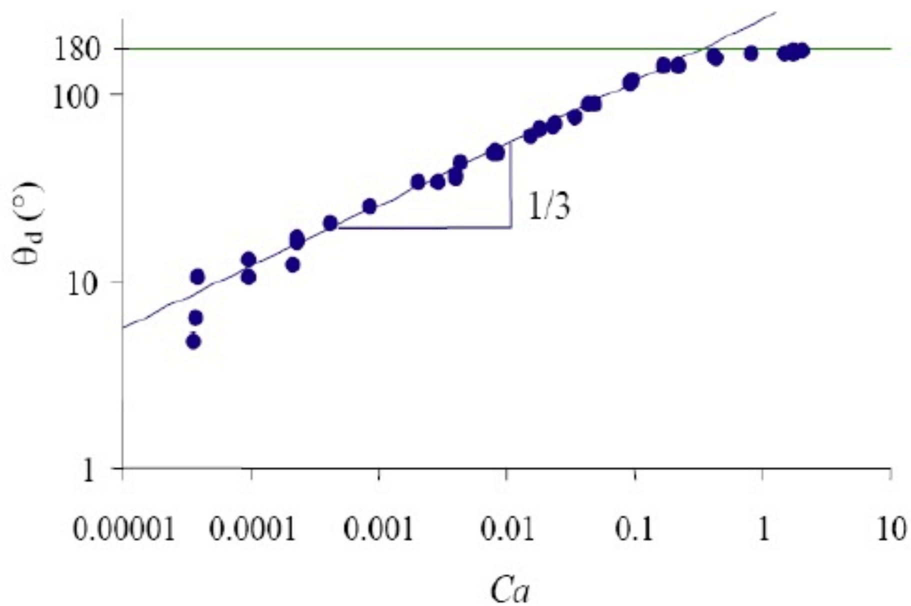


Dynamic Capillary effects



In composite processing, wetting is dynamic, the front has a given velocity…
And the contact angle varies with the local velocity:

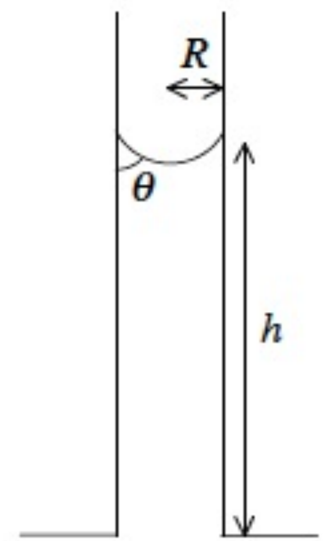
$$\theta^3 - \theta_0^3 \propto Ca, with \quad Ca = \frac{\eta . v_l}{\gamma_{ma}}$$



Hoffman, 1975, wetting liquid in a dry tube

Capillary pressure drop

Replacing a solid/air interface by a solid/liquid interface



In a cylindrical tube : $$\Delta P_{\gamma} = \frac{-2\gamma_{LV} \cos\theta}{R}$$

For an assembly of tubes or porous medium, there is a thermodynamic value of the capillary pressure as well:

$$\Delta P_{\gamma} = -\gamma_{LV} S_f \cos\theta$$

Where S_f is the interface area per volume of resin

If this pressure drop is negative, the system is wetting and impregnation is spontaneous.

For most polymers, low surface tension, whatever the contact angle, the capillary pressure is low.

Surface tension values

Material	Temperature (°C)	Surface Tension (N/m)
Polypropylene (PP)	180	0.0208
Polyethylene (PE)	180	0.0265
Polyethylene oxide (PEO)	180	0.0307
Nylon 6.6	270	0.0303
PEI	220	0.0357
PA 12	-	0.039
Epoxy, unreacted	-	0.03 to 0.04
Ethanol	20	0.022
Water	20	0.073
SiO_2	1800	0.31
Na_2SiO_3	1088	0.30
Al_2O_3	2050	0.63
$CaSiO_3$	1540	0.35
Al	700	0.87
Cu	1120	1.2
Ti	1670	1.53
Ag	970	0.92
Au	1070	1.13

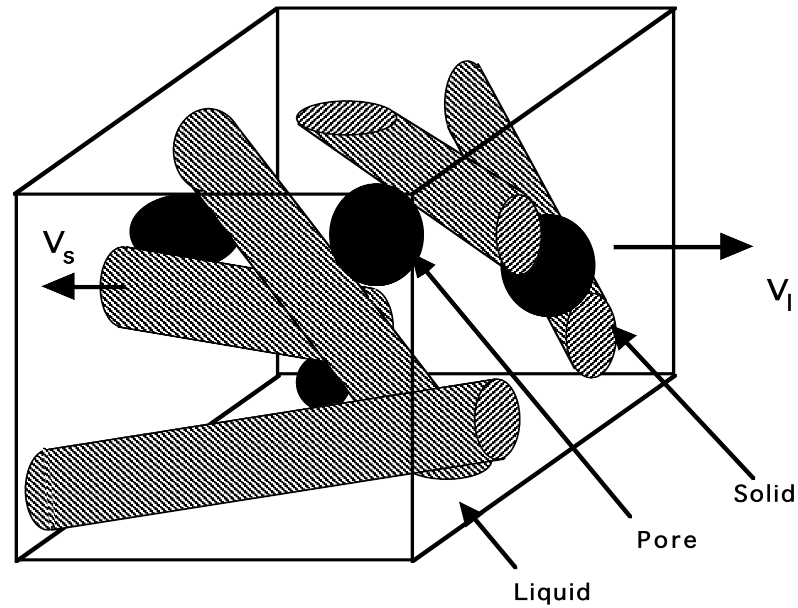
Infiltration modelling: Representative volume element

Homogenization technique: volume averaged quantities

$$V_l + V_p + V_f = 1$$

and

$$S = \frac{V_l}{(1 - V_f)}$$

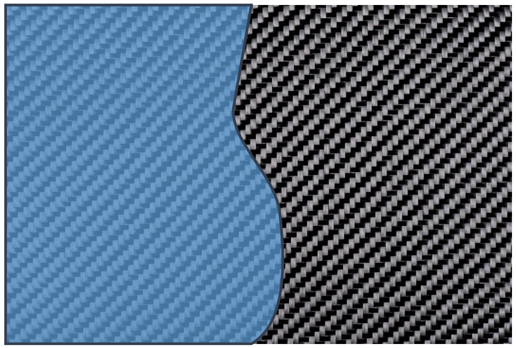


The Representative volume element (REV):
small on the scale of the preform
large on the scale of individual fibers

Resin Flow

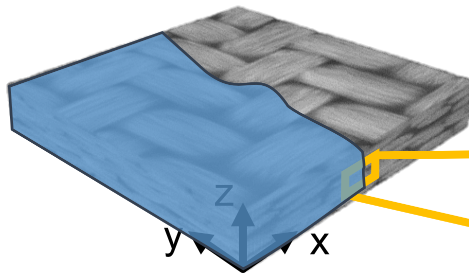
Other assumptions

- Isothermal (curing starts after injection)
- Stationary fibers
- Newtonian fluid



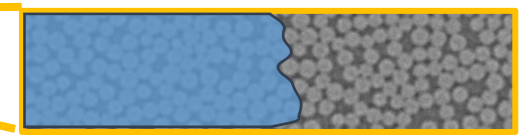
Woven fabric

(~ 1 – 10 m)



Yarns

(~ 1 – 10 mm)



Fibers

(~ 10 – 50 μm)

Conservation of mass

$$\frac{\partial \rho}{\partial t} + \frac{\partial}{\partial x}(\rho u_x) + \frac{\partial}{\partial y}(\rho u_y) + \frac{\partial}{\partial z}(\rho u_z) = -s$$

ρ = density [*kg/m³*]

u_i = velocity component in *i*-direction *[m/s]*

s = source/sink term [*kg/(m³.s)*]

Conservation of momentum

$$\rho\left(\frac{\partial u_x}{\partial t} + u_x\frac{\partial u_x}{\partial x} + u_y\frac{\partial u_x}{\partial y} + u_z\frac{\partial u_x}{\partial z}\right) = -\frac{\partial P}{\partial x} + \eta\left[\frac{\partial^2 u_x}{\partial x^2} + \frac{\partial^2 u_x}{\partial y^2} + \frac{\partial^2 u_x}{\partial z^2}\right] + F_{Bx}$$

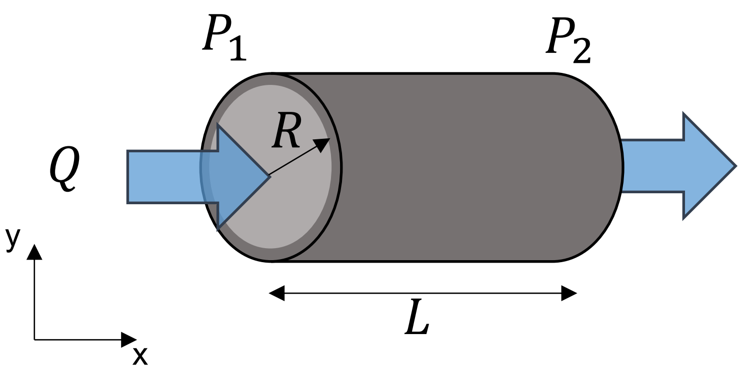
$$\rho\left(\frac{\partial u_y}{\partial t} + u_x\frac{\partial u_y}{\partial x} + u_y\frac{\partial u_y}{\partial y} + u_z\frac{\partial u_y}{\partial z}\right) = -\frac{\partial P}{\partial y} + \eta\left[\frac{\partial^2 u_y}{\partial x^2} + \frac{\partial^2 u_y}{\partial y^2} + \frac{\partial^2 u_y}{\partial z^2}\right] + F_{By}$$

$$\rho\left(\frac{\partial u_z}{\partial t} + u_x\frac{\partial u_z}{\partial x} + u_y\frac{\partial u_z}{\partial y} + u_z\frac{\partial u_z}{\partial z}\right) = -\frac{\partial P}{\partial z} + \eta\left[\frac{\partial^2 u_z}{\partial x^2} + \frac{\partial^2 u_z}{\partial y^2} + \frac{\partial^2 u_z}{\partial z^2}\right] + F_{Bz}.$$

Resin Flow

Hagen-Poiseuille equation

laminar flow of an incompressible fluid in a long cylindrical pipe with constant cross section



$$Q = \frac{-(P_2 - P_1)}{L}\frac{\pi R^4}{8\eta}$$

Q = flow rate [m^3/s]

$P_{1,2}$ = fluid pressure [*Pa*]

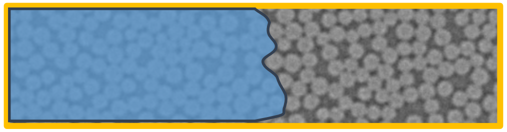
L = tube length [*m*]

R = tube radius [*m*]

η = viscosity [*Pa.s*]

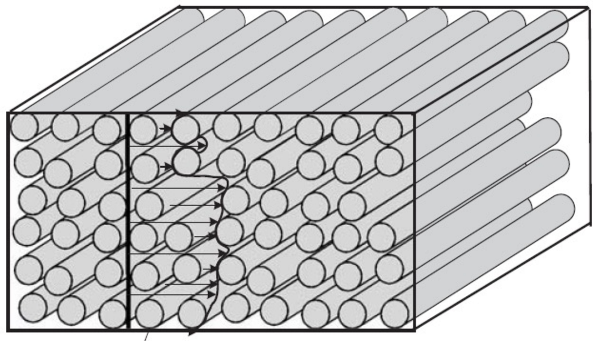
Other assumptions

- Isothermal (curing starts after injection)
- Stationary fibers
- Newtonian fluid



Fibers

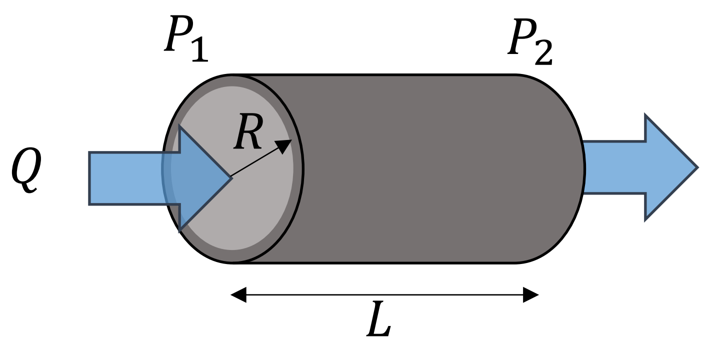
(~ 10 – 50 µm)



Resin Flow

Darcy's Law:
- Valid for laminar flow of incompressible fluids

Hagen-Poiseuille equation



$$Q = \frac{-(P_2 - P_1)}{L}\frac{\pi R^4}{8\eta} = \frac{\pi R^4}{8\eta}\frac{dP}{dx}$$

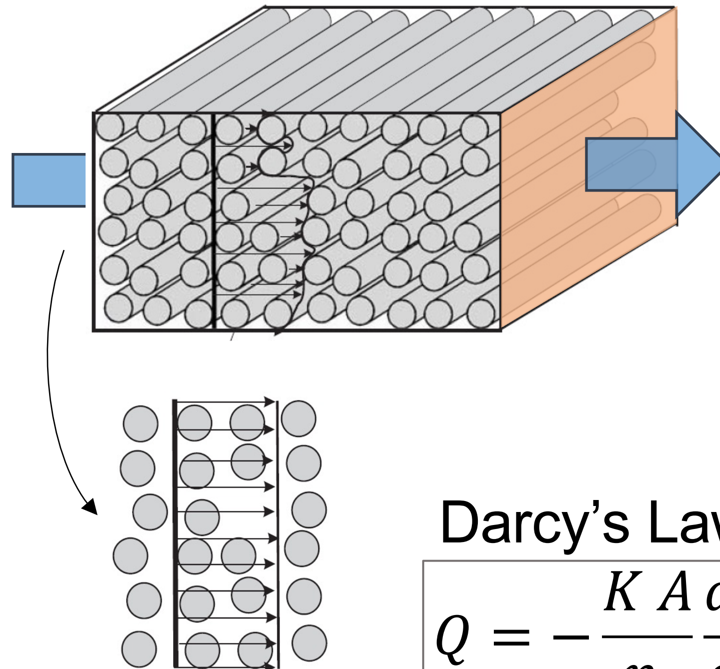
Q = flow rate [m^3/s]

$P_{1,2}$ = fluid pressure [*Pa*]

L = tube length [*m*]

R = tube radius [*m*]

η = viscosity [*Pa.s*]



Darcy's Law

$$Q = -\frac{K\,A}{\eta}\frac{dP}{dx}$$

K = permeability [m^2]: ease of flow through a porous medium

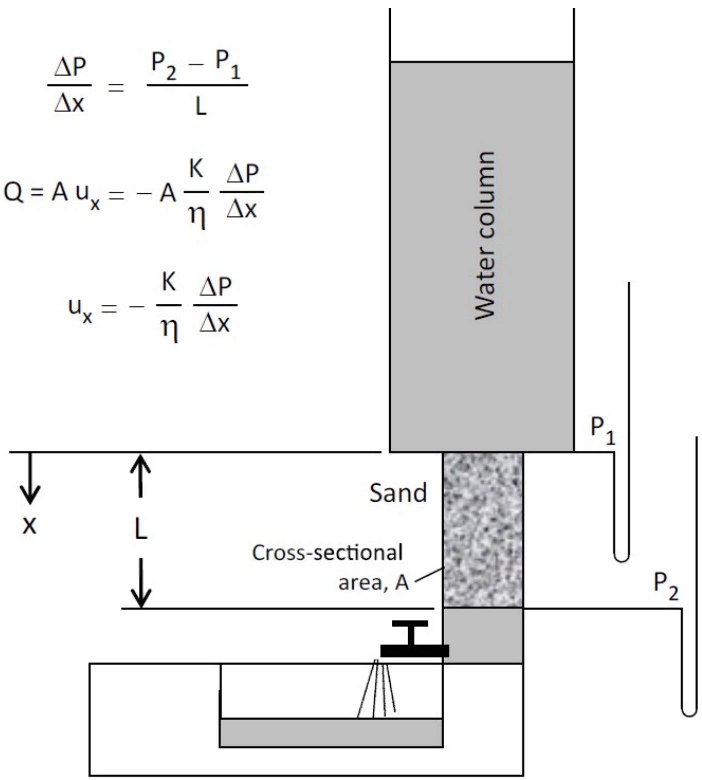
A = cross-sectional area [m^2]

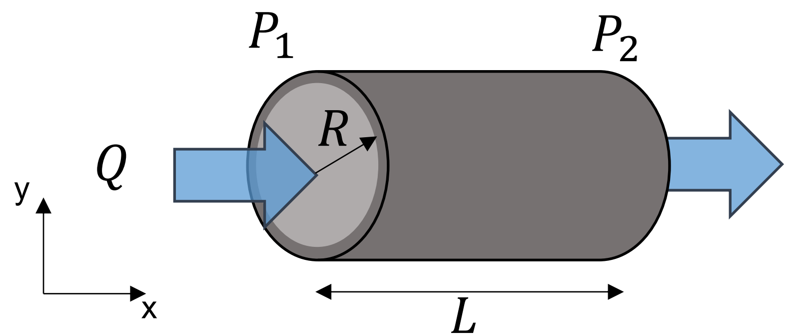
Darcy's law

$$\frac{\Delta P}{\Delta x} = \frac{P_2 - P_1}{L}$$

$$Q = A\,u_x = -A\,\frac{K}{\eta}\,\frac{\Delta P}{\Delta x}$$

$$u_x = -\frac{K}{\eta}\,\frac{\Delta P}{\Delta x}$$





Hagen-Poiseuille equation

$$Q = \frac{\pi R^4}{8\eta}\frac{dP}{dx} \longrightarrow Q = -\frac{R^2}{8}\frac{A}{\eta}\frac{dP}{dx}$$

Darcy's Law

$$Q = -\frac{K\,A}{\eta}\frac{dP}{dx}$$

Q = flow rate [m^3/s] η = viscosity [$Pa.s$] A = cross-sectional area [m^2]

P = fluid pressure [Pa] K = permeability [m^2] R = tube radius [m]

Energy conservation

Internal energy: $u=h-p/\rho$

$$\partial_t(\rho_\alpha . h_\alpha) + \nabla(\rho_\alpha . h_\alpha v_\alpha) - \partial_t p_\alpha - v_\alpha . \nabla p_\alpha + \nabla q_\alpha = 0$$

Phase α Density ρ_α Specific enthalpy h_α

Velocity $\mathbf{v}_\alpha$ pressure $\boldsymbol{p}_\alpha$ Heat flux $\mathbf{q}_\alpha$

Written in temperature:

$$\nabla(k_c \nabla T) = \rho_c c_c \frac{\partial T}{\partial t} + \rho_l c_l v_l . \nabla T + \rho_l \Delta H$$

Stress equilibrium

$$\sigma = \sigma' + BSp$$

σ applied stress tensor, σ' effective stress tensor (positive in compression), B Biot matrix, S saturation and p is local pressure in the fluid.
if isotropic, B=b I, where :

$$b = 1 - \frac{C_0}{C_s}$$

With C_0 the compression modulus of the fiber bed, and C_s the intrinsic compression modulus of the fiber material.

Terzhagi (usual assumption if reinforcement is hard): b=1

Mechanical equilibrium in practice

$$\sigma = \sigma' + Sp\begin{pmatrix} a_{11} & 0 & 0 \\ 0 & a_{22} & 0 \\ 0 & 0 & a_{33} \end{pmatrix}$$

In general, we neglect effects in the directions x and y in plane.

In the transverse direction:

$$\sigma_{zz} = \sigma_{zz}' + Sp$$

Summary Governing equations, in 1D

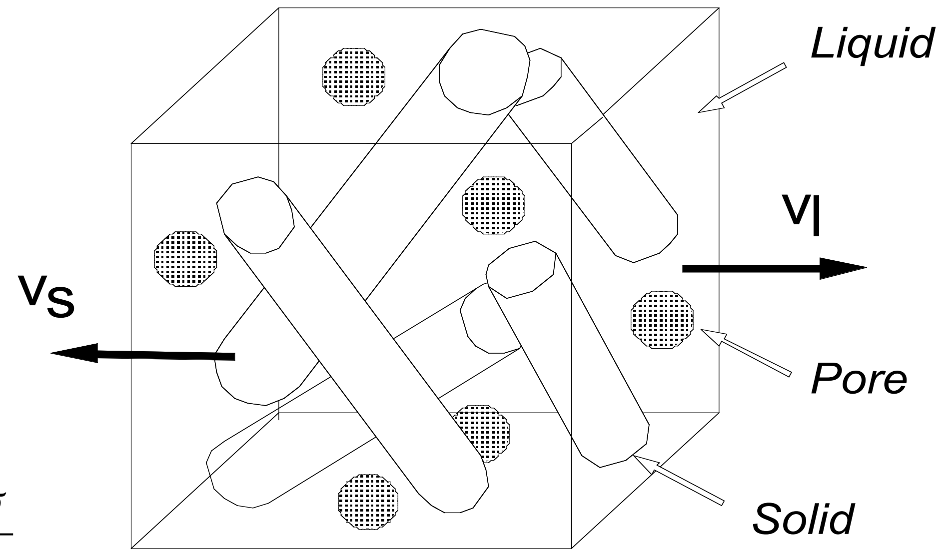
- Darcy's law: $$\left(1-V_f\right)S\left(v_l - v_s\right) = -\frac{K}{\eta}\frac{\partial P}{\partial x}$$

- Mass Conservation: $$\frac{\partial V_f}{\partial t} + \frac{\partial\left(V_f u_s\right)}{\partial x} = 0$$

and: $$\frac{\partial\left((1-V_f)S\right)}{\partial t} + \frac{d}{dx}\left(\left(1-V_f\right)S v_l\right) = 0$$

- Mechanical equilibrium: $$\frac{\partial(SP)}{\partial x} = -\frac{\partial \sigma}{\partial x}$$

With the saturation: $$S = \frac{V_l}{1-V_f}$$



Usual assumptions

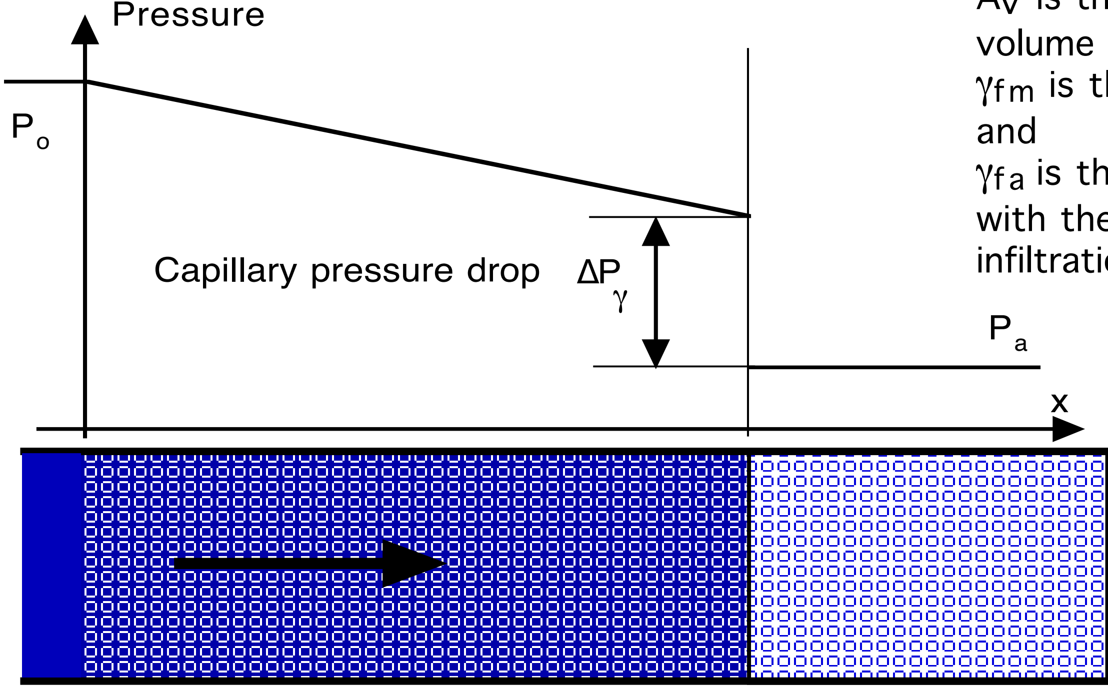
- non compressible fluid and solid phases
- laminar flow
- no phase change (solidification)
- isothermal

Boundary conditions

- Geometry of the part, symmetry, etc…
- Temperature
- Pressure applied at the resin entrance, or imposed flow rate
- Pressure or stress imposed on the assembly

- Pressure at the infiltration front ?
  - Two strategies:
    - multiphase flow, P =0 at the front, but Saturation curves
    - assume S=1 everywhere in the infiltrated part, saturated flow, and $P=\Delta P_{\gamma}$ at the front.

S=1, Slug flow assumption

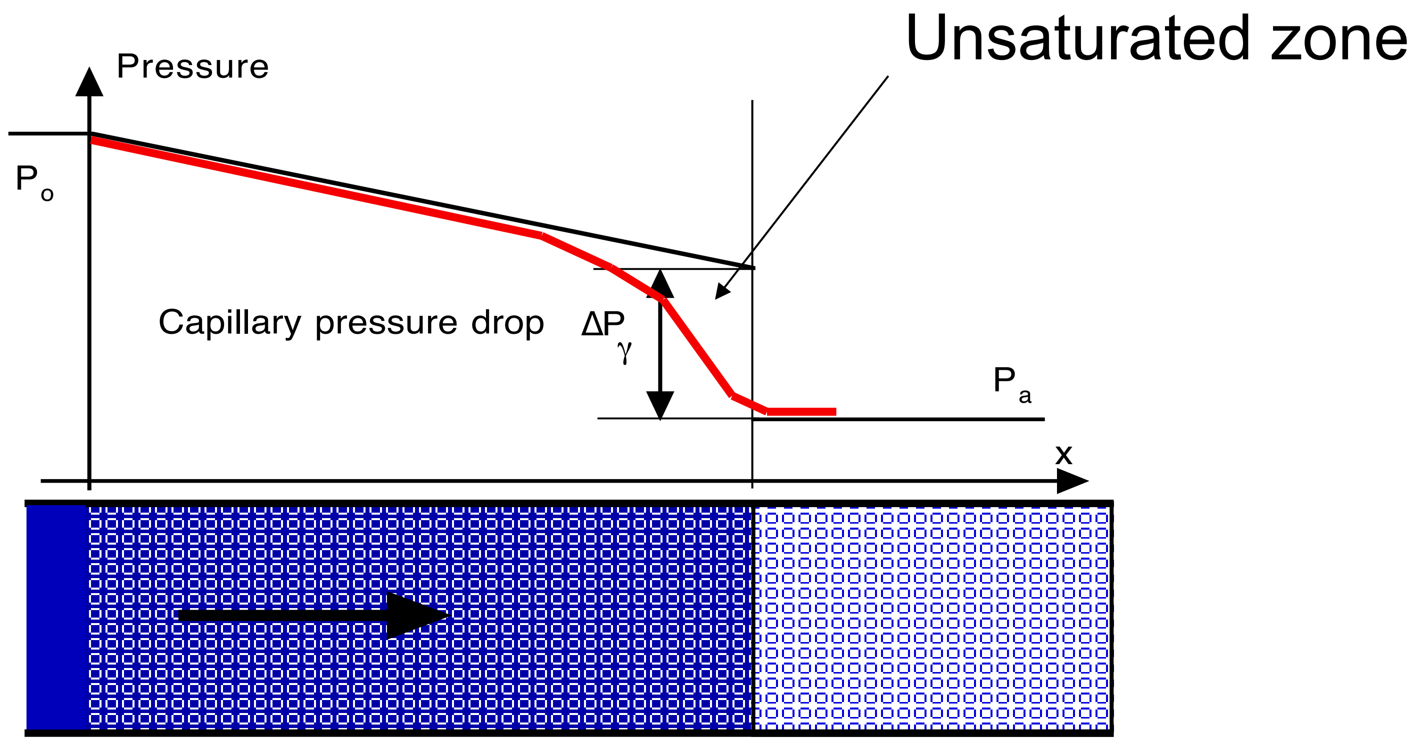


From a simple energy balance:

$\Delta P_\gamma = (1 - V_f)^{-1} A_V (\gamma_{fm} - \gamma_{fa})$,

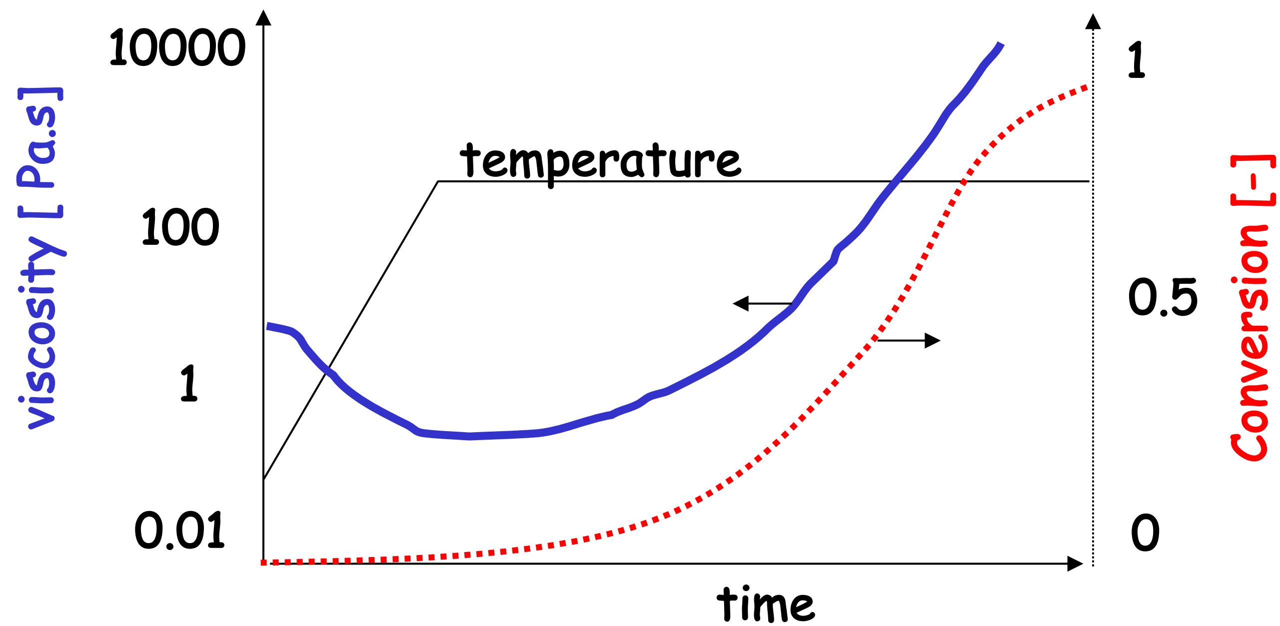
where
A_V is the area of matrix/reinforcement interface per unit volume of composite,
γ_{fm} is the matrix reinforcement interface surface energy, and
γ_{fa} is the surface energy of the reinforcement in contact with the initial atmosphere within its pores before infiltration (air, vacuum, etc)

S varies, multi-phase flow

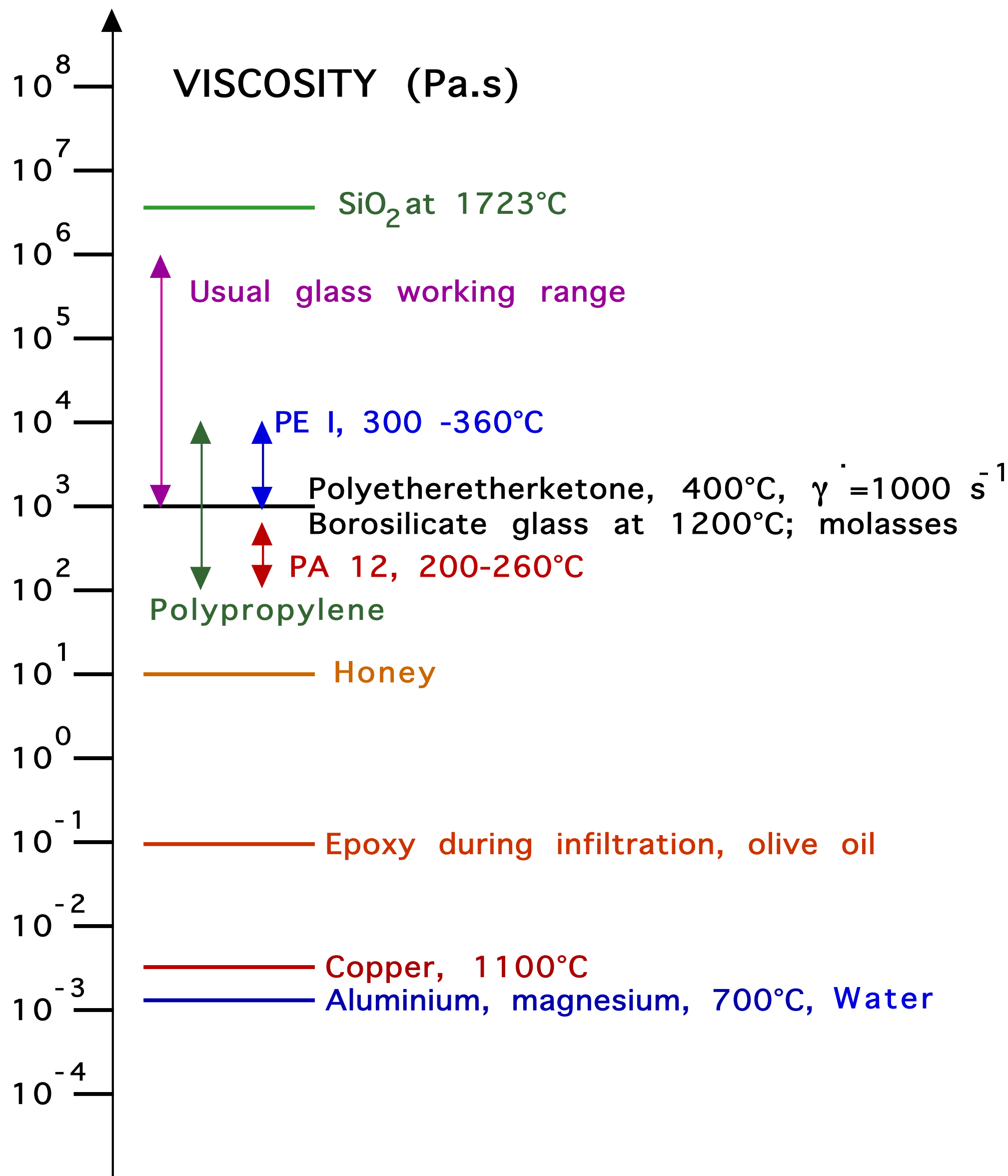


Parameters needed to solve the equations

Resin: viscosity, cure kinetics…



Matrix viscosity, η



Permeability

Permeability: a tensor, units m^2

$K=K_{sat}.\ K_r$
K_{sat} is the permeability to saturated flow, a function of the fibre bed architecture and volume fraction only, tensor

K_r is the relative permeability, function of the saturation S

Saturated Permeability prediction

Flow in a single tube:

$$u_x(r) = -\frac{\partial p}{\partial x}\frac{R^2}{4\eta}\left[1-\left(\frac{r}{R}\right)^2\right]$$

$$Q = \left(\frac{-dp/dx}{2\eta}\right)\left(\frac{\pi R^4}{4}\right)$$

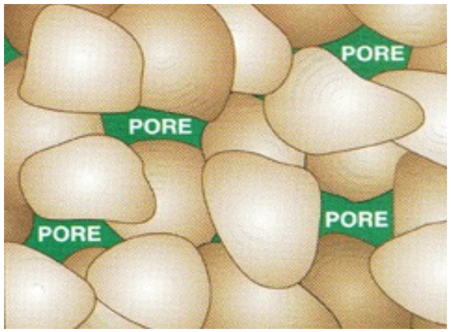
As mentioned earlier we find: $K=R^2/8$
Permeability is given in m^2

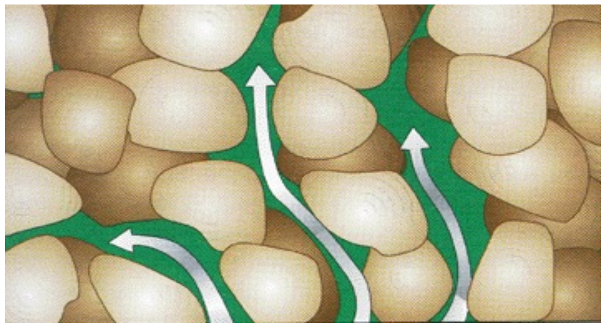
What is permeability of a porous medium?

Soil science

Permeability - represents the ability of a porous material to transmit fluids

Interconnected porosity

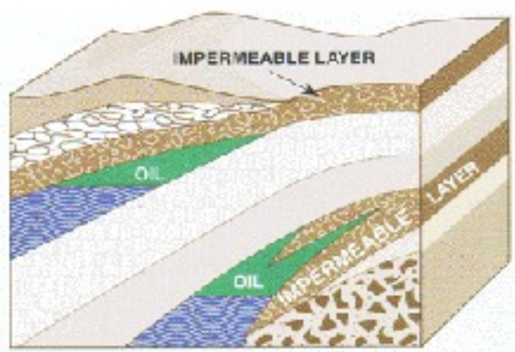


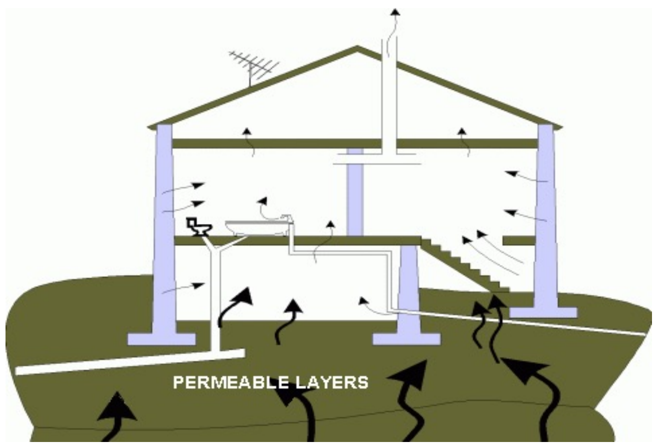


Darcy's law (1D):

$$v = -\frac{K}{\mu}\frac{\Delta P}{L}$$

v – superficial velocity
K – permeability
μ – viscosity
ΔP – pressure gradient
L – length





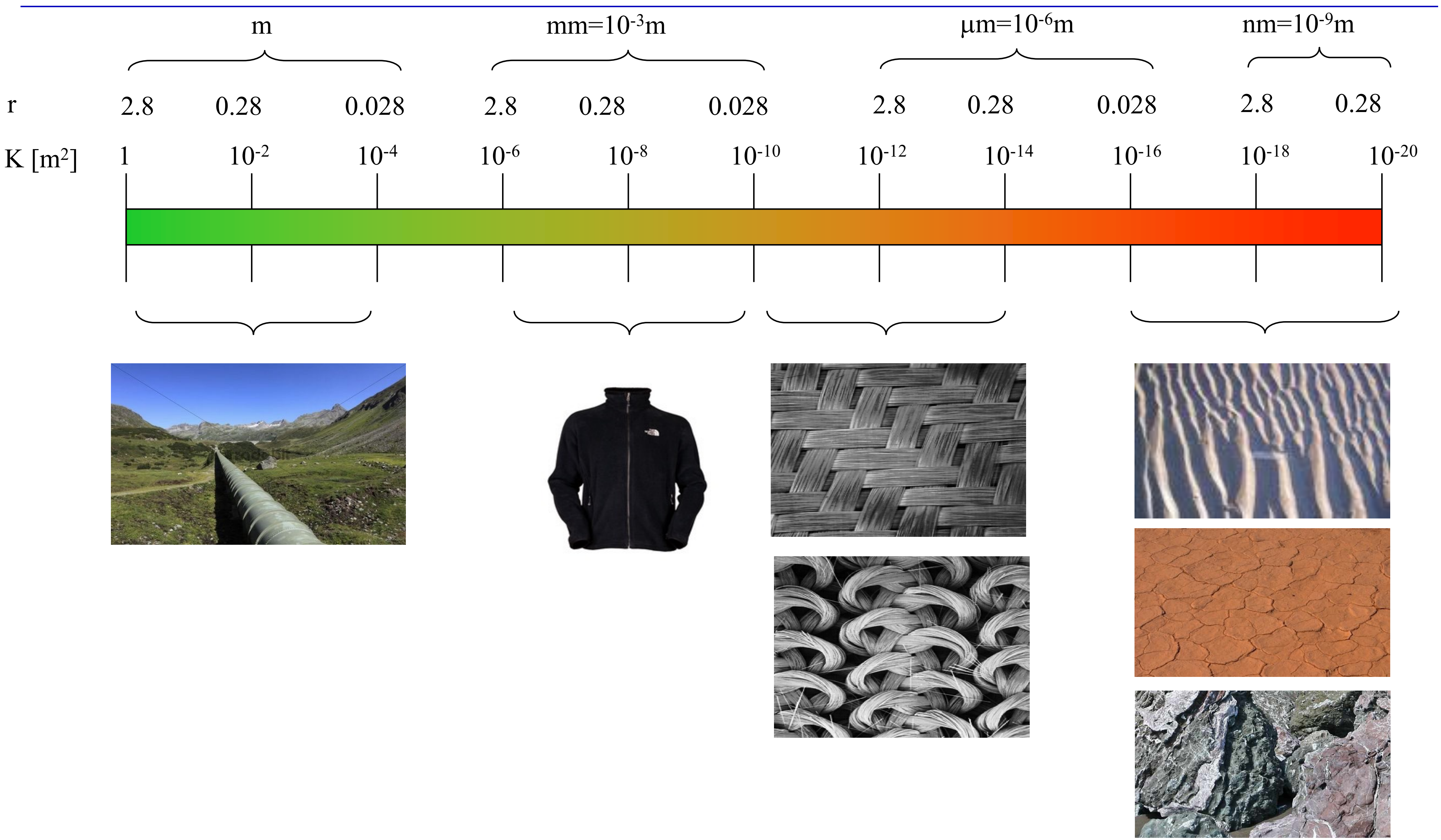
Analogy with Poiseuille's equation

$$\frac{Q}{A} = \frac{1}{\mu}\frac{r^2}{8}\frac{\Delta P}{L}$$

$$u_x(r) = -\frac{\partial p}{\partial x}\frac{R^2}{4\eta}\left[1-\left(\frac{r}{R}\right)^2\right]$$

S. Tavares

Permeability range



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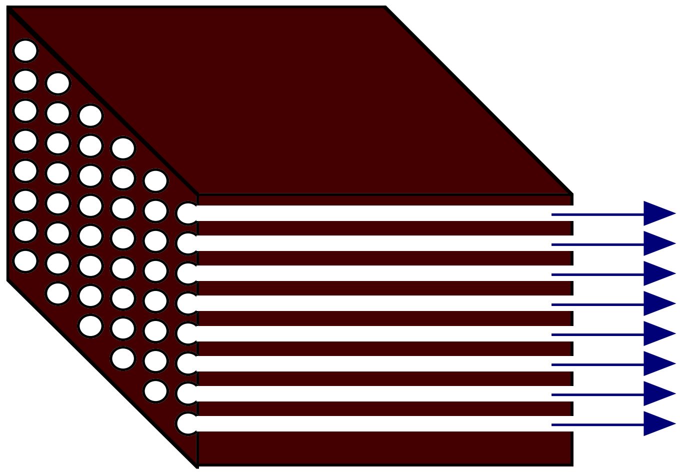
Saturated Permeability prediction

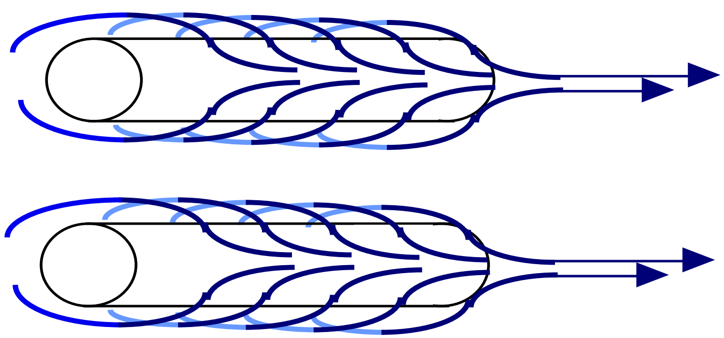


Parallel tube bundle model: Carmann-Kozeny equation (Carman 1938, ...)

$$K = \frac{R^2}{4k_{i,i}} \frac{(1-V_f)^3}{V_f^2}$$

R is the fiber radius, V_f is the fiber volume fraction, $k_{i,i}$ the Kozeny-Carman constant (i=x, y, z)

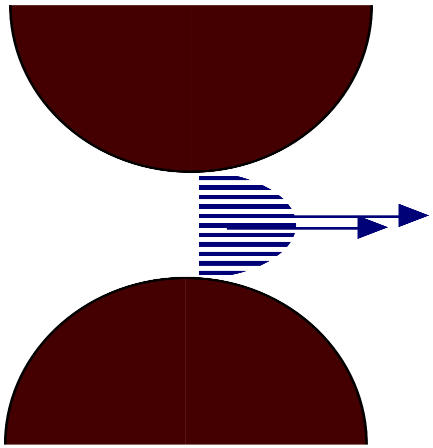
$$\frac{K}{r^2} = \frac{1}{8\Phi}\left(2\Phi - Ln(\Phi) - 1.476 - 1.774\Phi + 4.076\Phi^3\right)$$



Viscous drag models (Langmuir 1942, Happel 1959, ...)

Φ est la fraction volumique de fibres et $\leq 10\%$, quadratic arrangement, r le rayon de la fibre

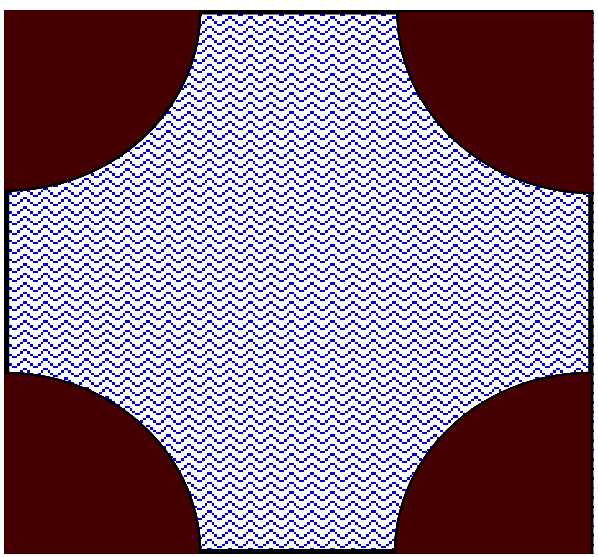
Saturated Permeability prediction



Lubrication theory
(Keller 1964 ...)

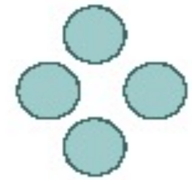
$$\frac{K}{r^2} = \frac{1}{9\Phi\sqrt{2}}\left(1-\sqrt{\frac{4\Phi}{\pi}}\right)^{5/2}$$

$10\% \leq \Phi \leq \Phi_{max}$, quadratic arrangement



Unit cell models
(Sangani and Acrivos 1982, Drummond and Tahir 1984...)

Saturated Permeability prediction



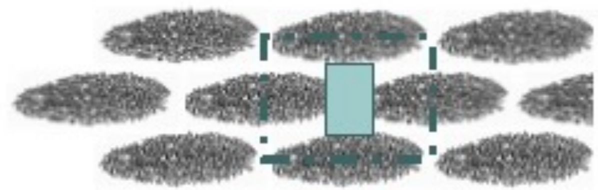
Gebart (1992)

$$K = \frac{16}{9\pi\sqrt{2}}\left[\sqrt{\frac{1-\phi_{\max}}{1-\phi}}-1\right]^{2.5} R^2$$

Avec $\phi_{\max} = 1-\frac{\pi}{2\sqrt{3}}$

Lundström (2000)

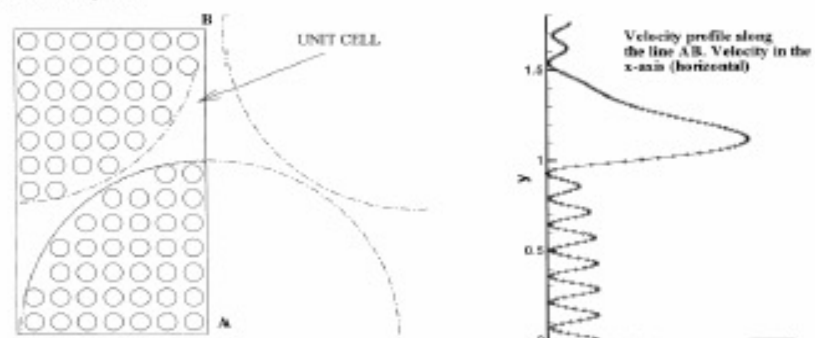
$$K = \frac{w^3}{h}\left[\frac{16}{\pi^5}\sum_{\alpha=0}^{\infty}-\frac{1}{\beta^5}\frac{\exp\left(\beta\pi\frac{h}{w}\right)-1}{\exp\left(\beta\pi\frac{h}{w}\right)+1}+\frac{1}{12}\frac{h}{w}\right]+\frac{K_{tow}}{1-Vf_{tow}}$$



Papathanasiou (2001)

$$K = K_M\left[1+2.67\left(\frac{K_{tow}}{K_M}\right)^{0.89}\right]$$

HEX-SQ arrangement

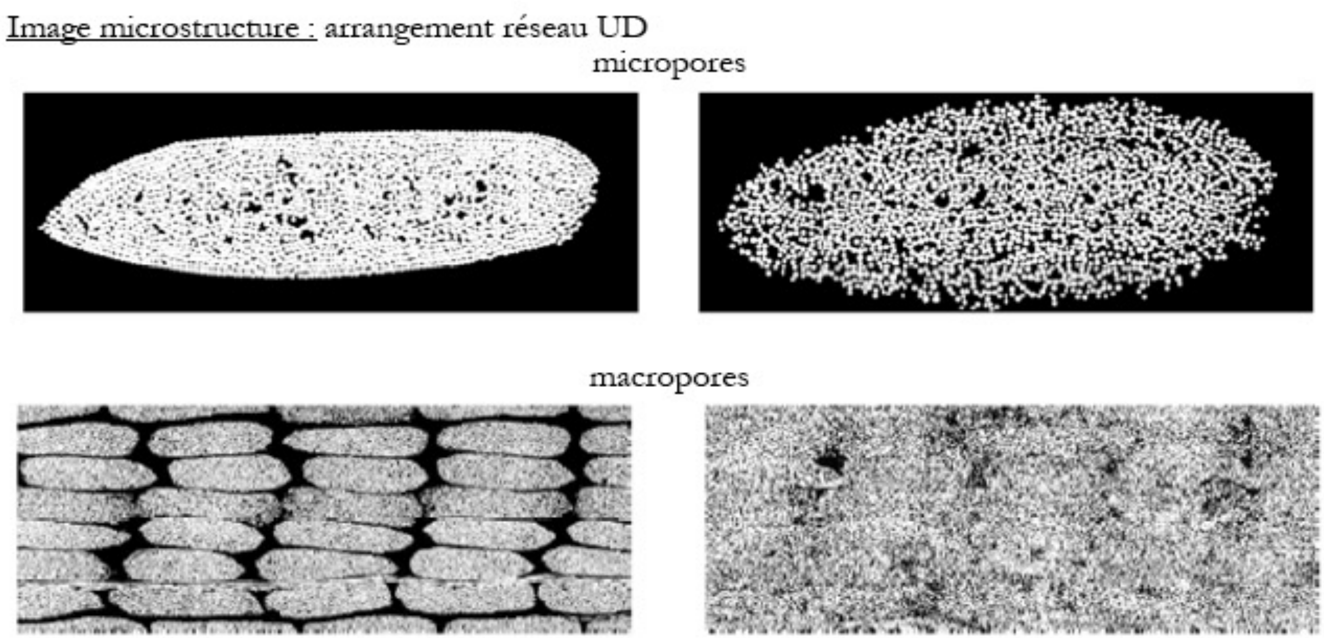


inter-tow porosity=12.5% intra-tow porosity=60% 148 fibers in a tow

(a) (b)

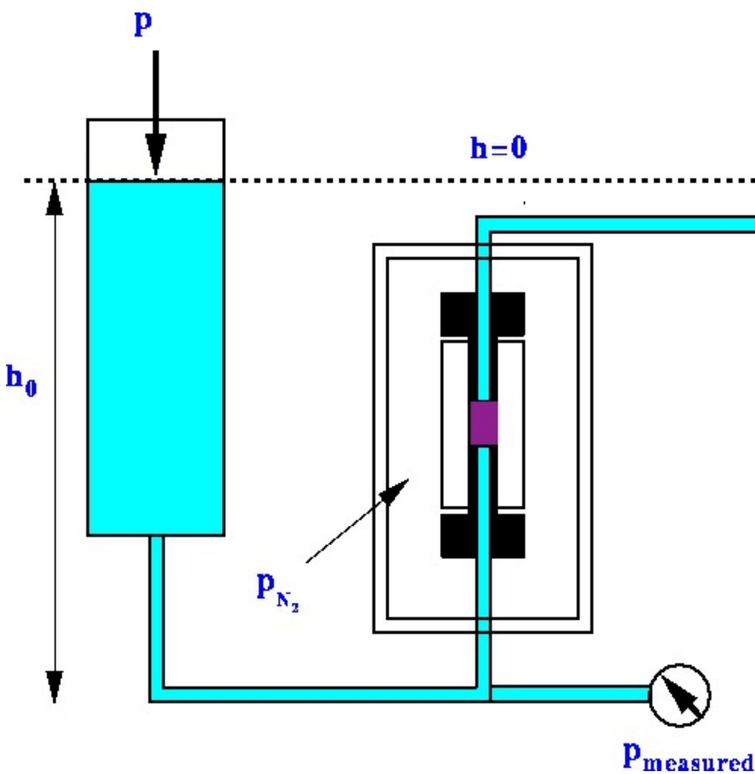
Saturated Permeability prediction

Unit cell models, reproducing the fiber architecture in a REV, then computational fluid dynamics to calculate the pressure drop-> permeability of the cell

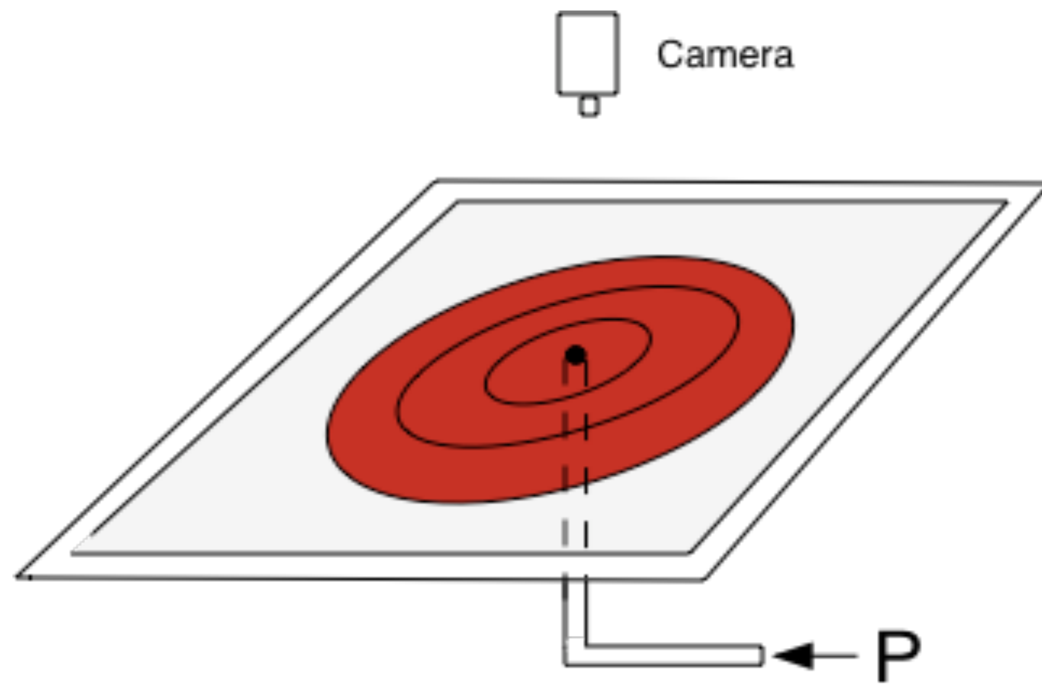


From Breard et al.

Saturated permeability measurement

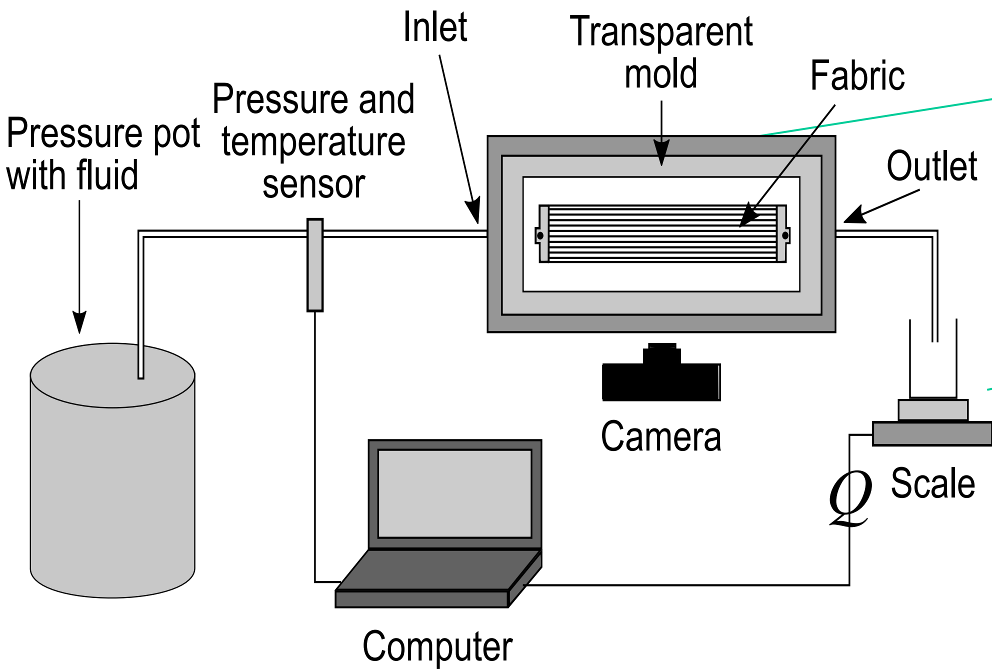


Transverse permeability



In-plane permeability

In plane permeability



$$K_{unsat} = \frac{-(1-V_f)\eta\psi^2}{2\Delta P}$$

$$K_{sat} = \frac{-Q\eta L_{tot}}{A\Delta P}$$

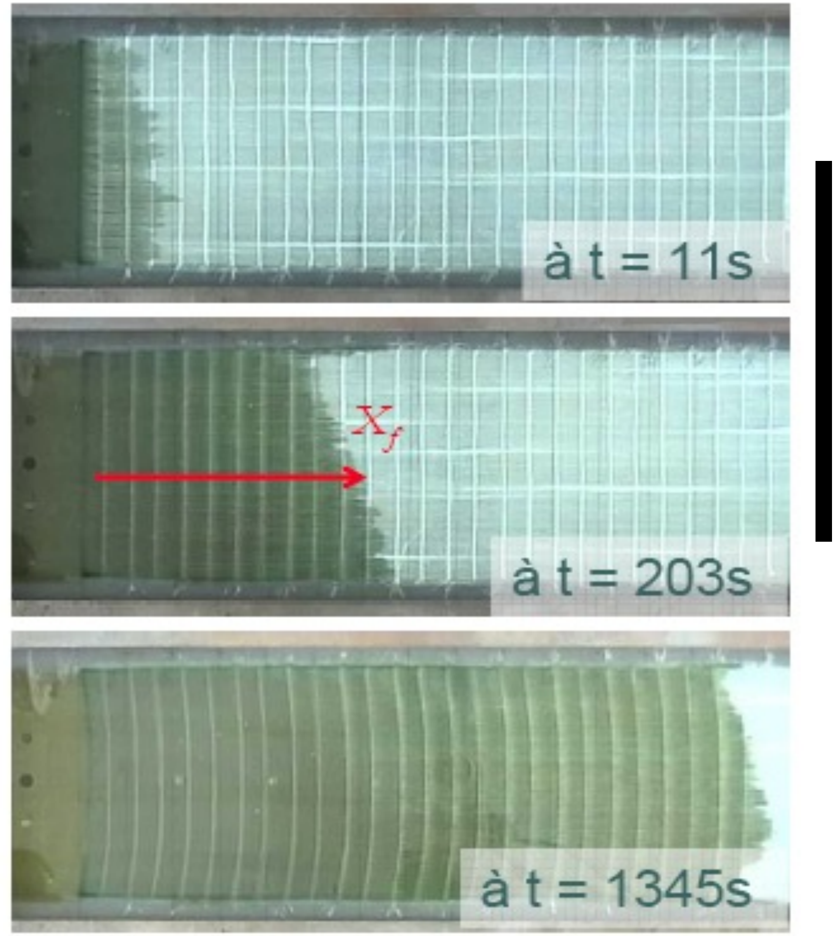
$L(t)$ = flow front position at time t

$-\Delta P$ = fluid pressure at inlet, Neglecting capillary pressure

Q = flow rate of outcoming fluid

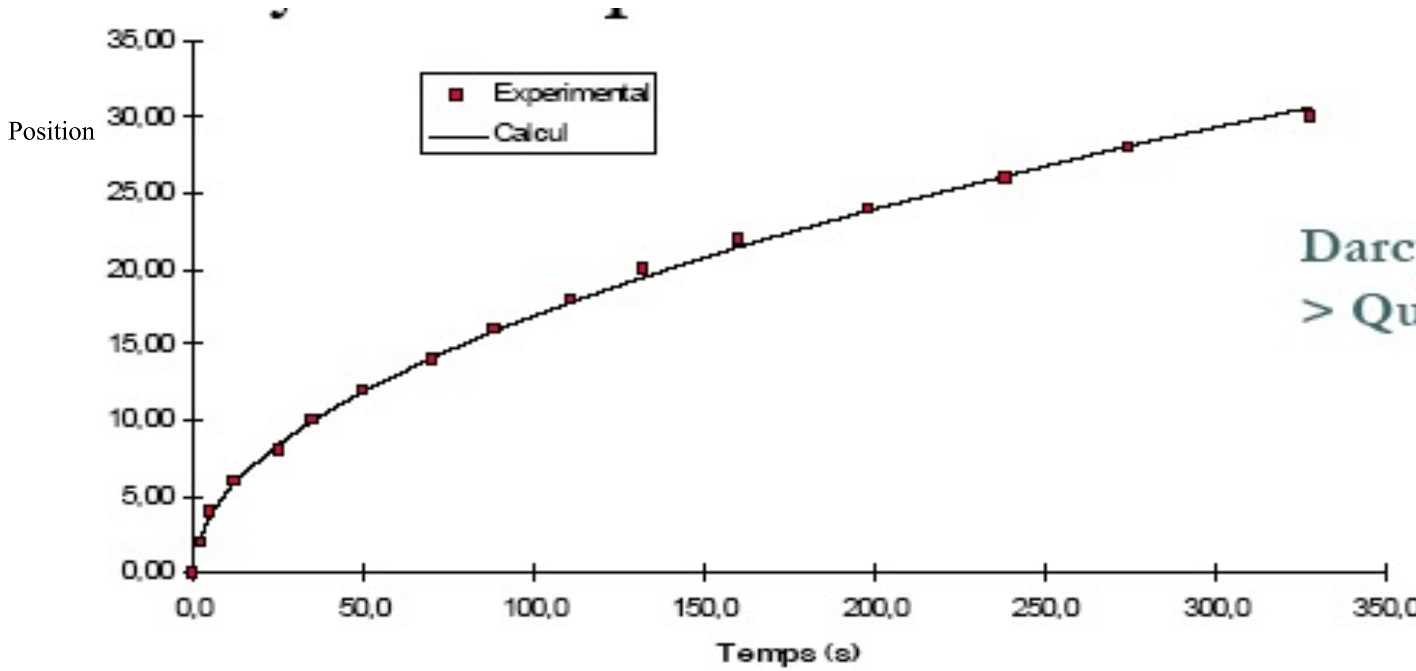
Permeability measurement

In-plane permeability



From Bréard et al,

Permeability measurement



Darcy
> Quasi-stationnaire

Stardard permeability measurement, plot flow front position versus time, evaluate $\psi = L^2/t$, use a fluid of well known viscosity, given applied pressure, evaluate K

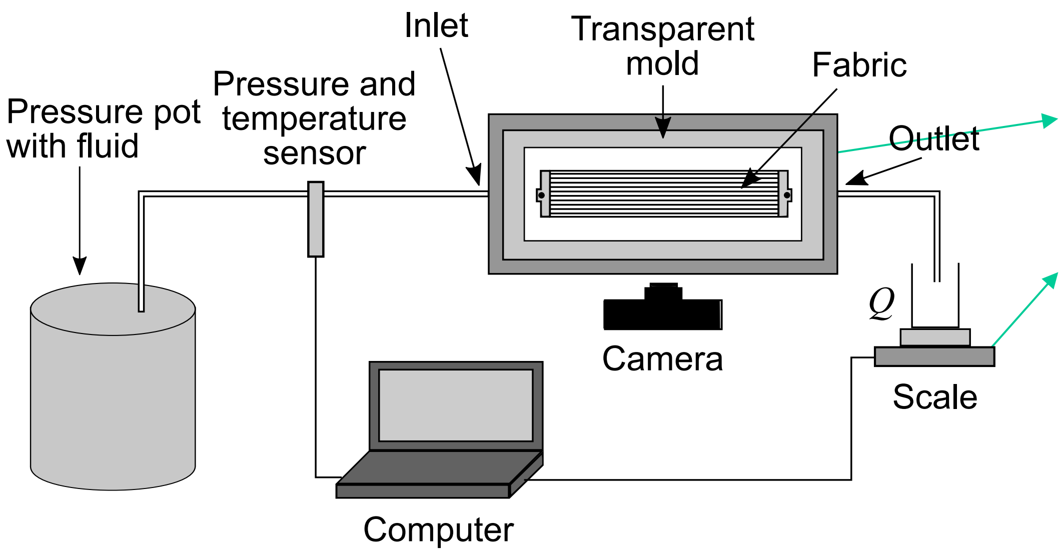
$$K_{unsat} = \frac{-(1-V_f)\eta\psi^2}{2\Delta P}$$

Attention, unsaturated permeability!!!!

From Bréard et al,

Saturated permeability measurement

Setup for **in-plane, constant pressure, unidirectional** permeability measurements, following the permeability RR guidelines, with a well known fluid, such as silicone oil.



$$K_{unsat} = \frac{-(1-V_f)\eta\psi^2}{2\Delta P}$$

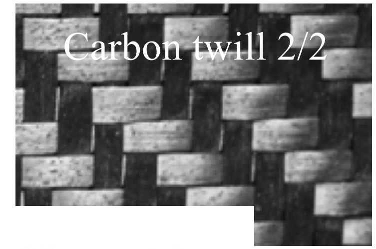
$$K_{sat} = \frac{-Q\eta L_{tot}}{A\Delta P}$$

K_{sat} and K_{unsat} are often not equal

$L(t)$ = flow front position at time t
$-\Delta P$ = fluid pressure at inlet, Neglecting capillary pressure
Q = flow rate of outcoming fluid

Permeability is not easy to measure in any case

Same fabric, Carbon Twill 2/2, sent to many laboratories



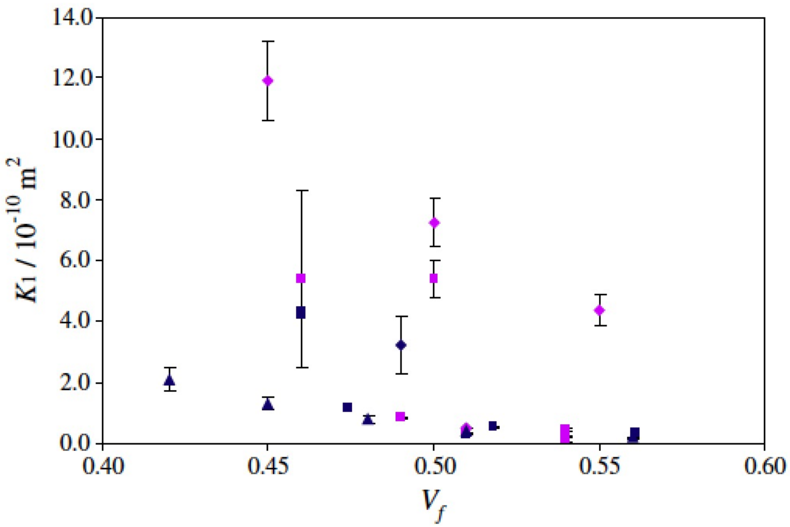
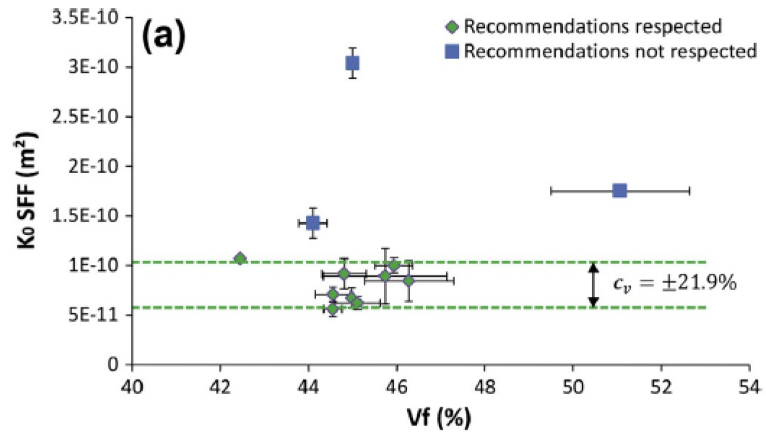


Fig. 1. G0986: Measured principal permeability value K_1 as a function of the fibre volume fraction V_f. Different symbols indicate experimental method (dark diamond: radial unsat.; light diamond: radial sat.; dark square: linear unsat.; light square: linear sat.; dark triangle: 1D compression).

R. Arbter et al. / Composites: Part A 42 (2011) 1157–1168



Permeability of the same fabric, measured following precise guidelines, K_{unsat}, silicone oil.

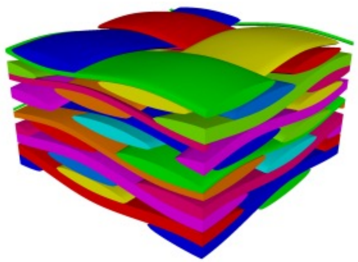
N. Vernet et al. / Composites: Part A 61 (2014) 172–184

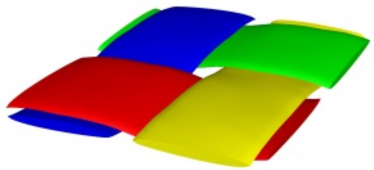
No precise guideline or standard ->A lot of scatter between various laboratory measurements of the same fabric. Precise guidelines…agreement but are the values correct?
Since 2023, there is an ISO norm on permeability measurement for in-plane. ISO 4410

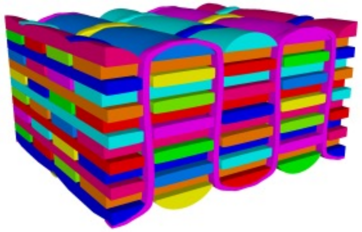
Preform stress-strain curve in a fluid

-During processing, preforms can be submitted to compression, and to shear and tensile forces in the plane.

- During composite processing, on can distinguish between the stresses applied by the mould before infiltration (shear, compression) and those that will be applied by the moving resin, which imply hydro-mechanical coupling in the equations.



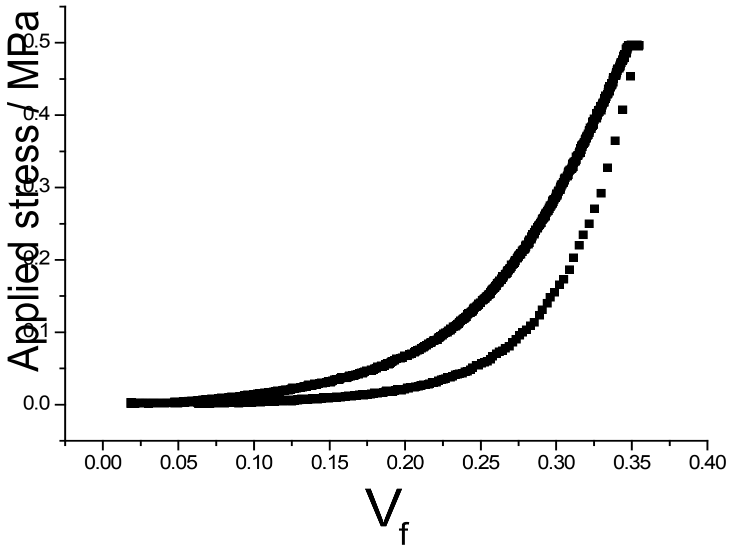




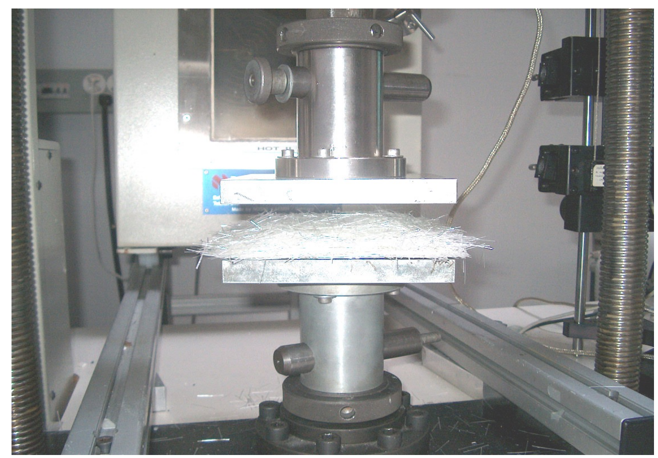
Stress-strain curves

- $\dfrac{d\sigma}{dV_f}$

- often non-linear
- hysteretic
- depends on the number of plies

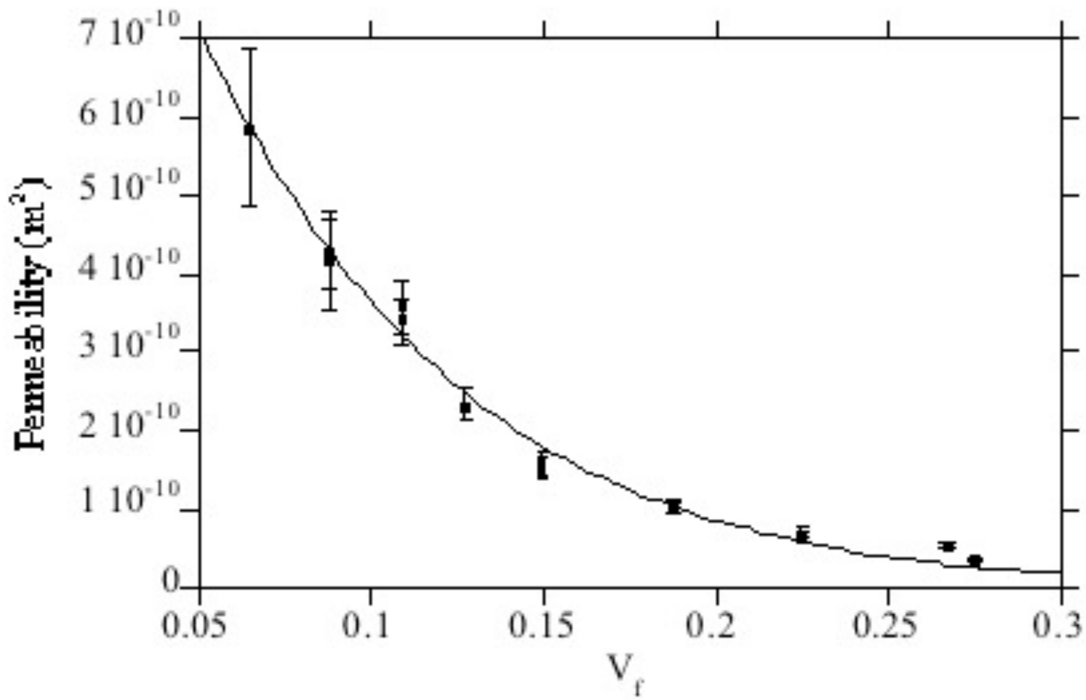


Compressive test



Permeability and compressibility Coupling

Example of GMT glass fibre mats



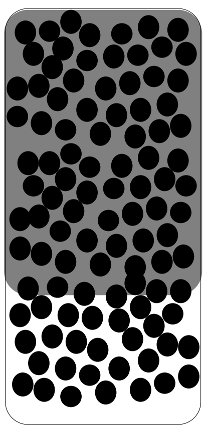
Practical cases of composite process modeling

Once the model is defined, and the parameters estimated, it is possible to scale -up to more complex geometries, or more complex cases of pressure increase, of heat transfer, etc..

-> use numerical tools, such as finite element softwares for fluid flow.

- at the level of a unit cell of textile
- at the level of the part

Solving the equations is the case of saturated unidirectional flow with constant applied pressure



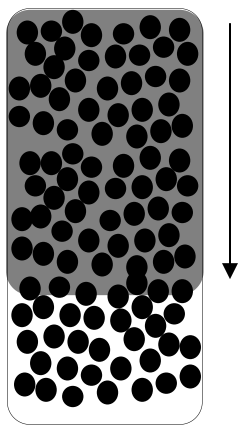
Exercise 1: Find the impregnation time, for saturated unidirectional infiltration of a fluid into a rigid fiber bed under constant applied pressure, for a given infiltration length L, as a function of the fluid viscosity, preform volume fraction and permeability, Applied pressure.

A simple case:UD infiltration, saturated , isothermal, rigid preform, under constant pressure

Mass conservation equation:

$$div(v_0) = 0$$

Momentum conservation equation (Darcy)



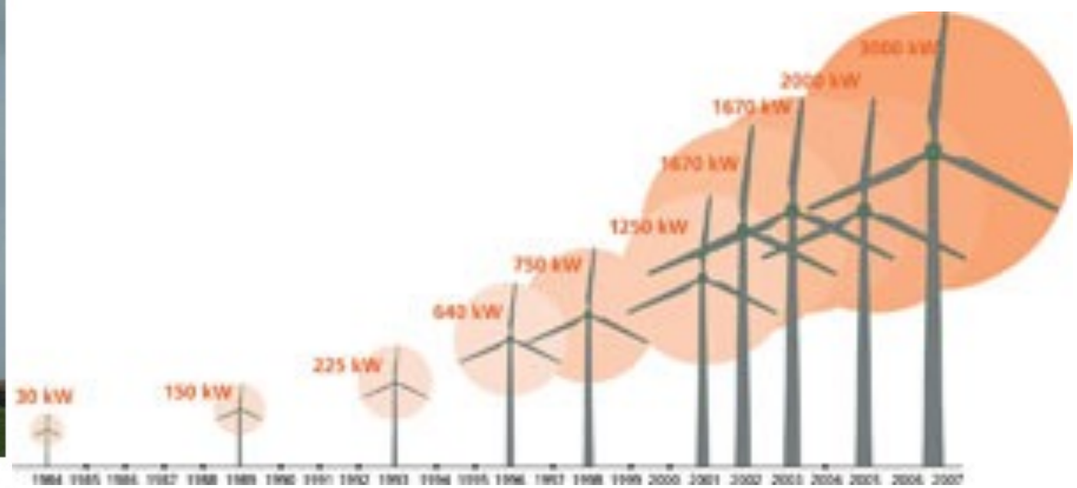
$$v_0 = \frac{Q}{A} = (1 - V_f)\frac{dL}{dt} = -\frac{K}{\eta}\frac{dP}{dx}$$

$$t_{\text{imprégnation}} = \eta \frac{(1 - V_f)L^2}{2K(P_a - \Delta P_c - P_{atm})}$$

A practical example: Windmill blade

Figure 1 The world's largest wind turbine (summer 2004) at Risoe National Laboratory test site, Høvsøre, Denmark. Tower height is 120 m, rotor diameter is 110 m, and generator power is 3.6 MW.

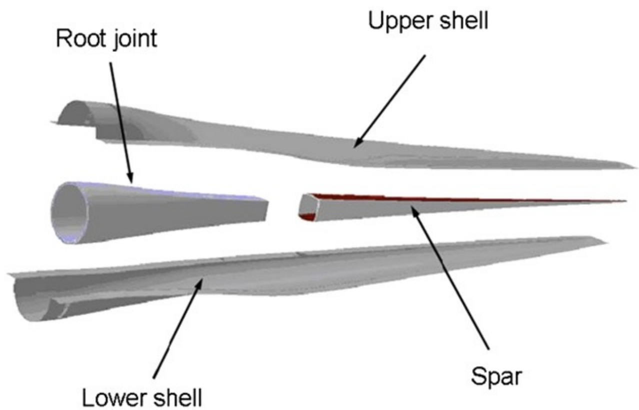
We need to produce a windmill blade, in composite: glass-vynil ester, PVC foam core.



Wind mill applications

Figure 1 The world's largest wind turbine (summer 2004) at Risoe National Laboratory test site, Høvsøre, Denmark. Tower height is 120 m, rotor diameter is 110 m, and generator power is 3.6 MW.

Aerodynamic blade design,
composite skins and spars, about 1500 parts per year



P.J. Schubel / Composites: Part B 43 (2012) 953–960

Main processing methods:

- Hand lay-up
- Prepreg technology in a female mould
- Resin infusion
- Other possibilities like filament winding

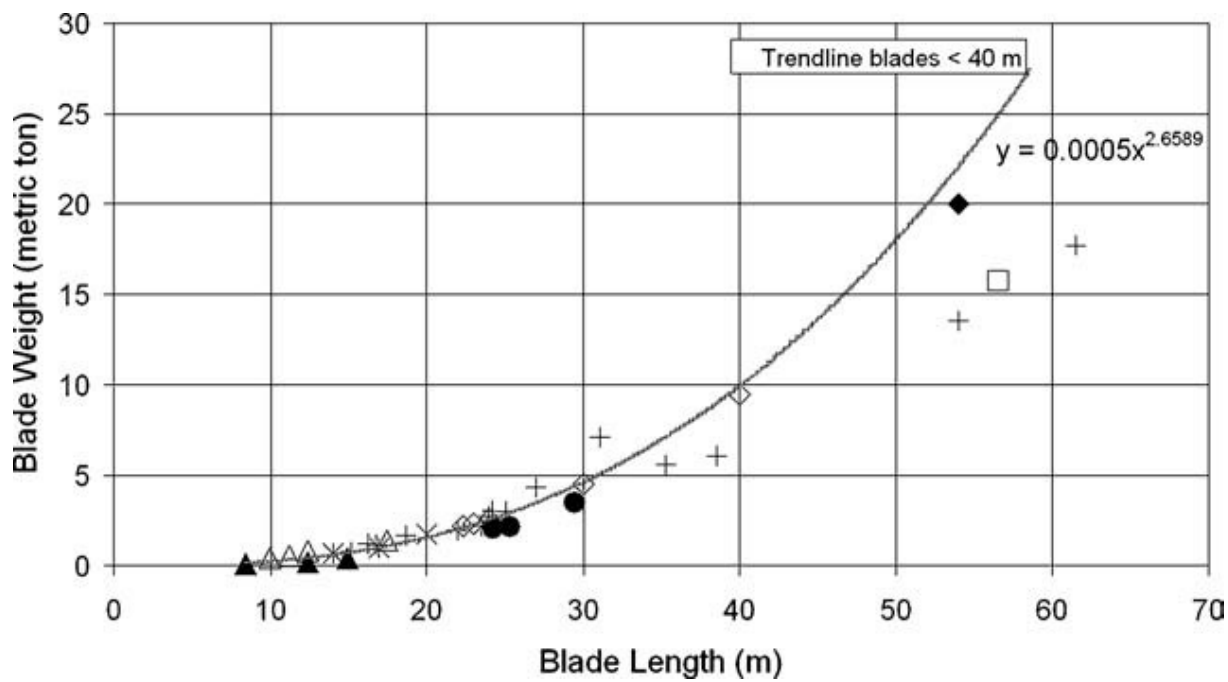
A possible technique

Vacuum infusion of resin

LM Glasfiber, Lunderskov, Denmark

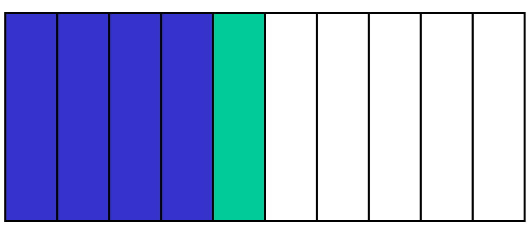
Exercise

-Could you evaluate the time to impregnate a blade, 40 m long?
Assume a flat panel, 40 m long, 2m wide, 3cm thick, 50% fibers, viscosity resin is 500mPas.

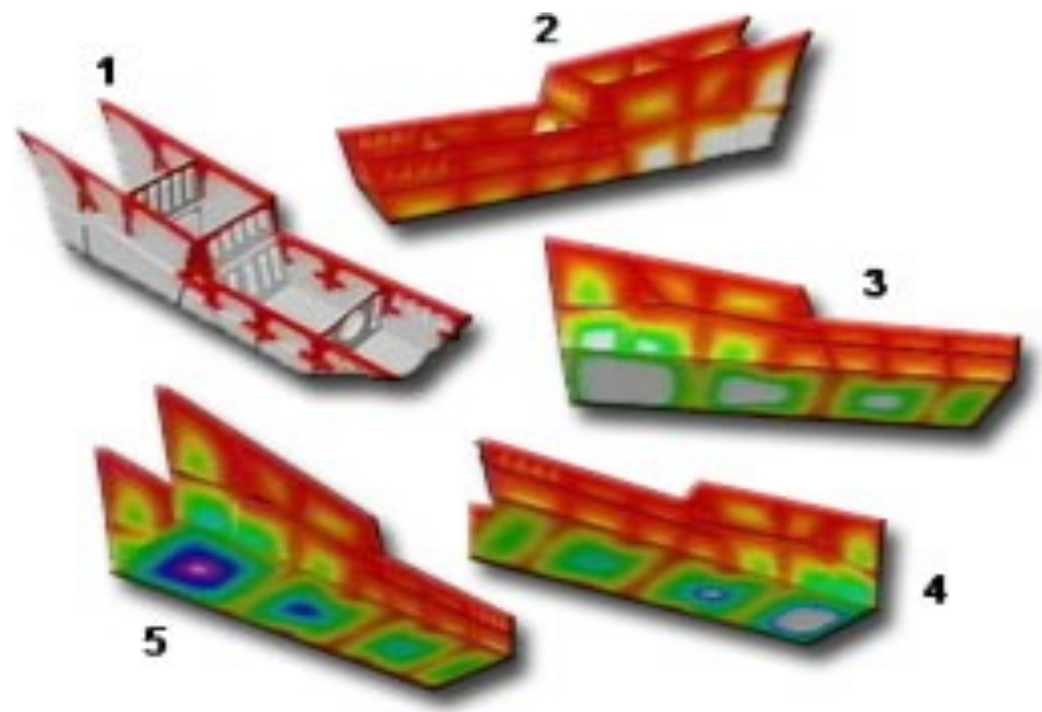


Practical cases , complex geometries

For FE modelling, problem of front tracking
vs mass conservation
- In 2, 3 D, finite element/control volume solution
 Exemple of LIMS (Delaware):



Elements filled



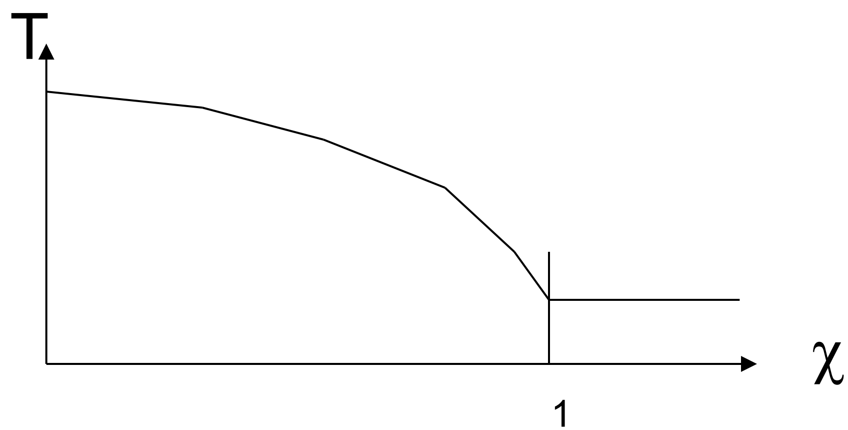
Influence of heat transfer?

In most cases, solve for HT after flow has ceased.

Heat transfer during flow, no chemical reaction or Solidification:

$$\nabla(k_c \nabla T) = \rho_c c_c \frac{\partial T}{\partial t} + \rho_l c_l v_l . \nabla T$$

This can be solved simultaneously with the other equations,
Even using the similarity solution for simple cases, constant P.
Example: cold preform, warmer resin, assume no conduction into the fibers ahead of the front



Remaining research issues

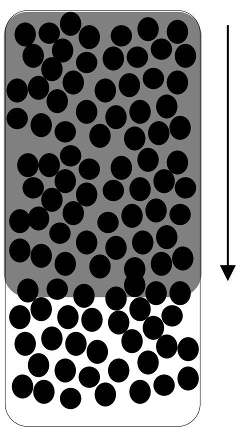
- Front tracking is still an issue in some cases, and no commercial software is yet able to predict potential porosity levels well (multiphase flow).

-Capillary effects are difficult to capture and model

- Permeability variation with shear, etc…needs to be taken into account

- Statistical nature of the process -> « intelligent manufacturing »

Conclusions



- Many processes exit for making composite materials, which can be classified into three major types.
-Process modelling is often performed using a Continuum mechanics approach, leading to a system of non-linear differential equations to solve.
- Need to make relevant assumptions
- Need to know and measure the constitutive equations or parameters to include in the model: stress-strain behaviour of the fiber bed, permeability, matrix rheology, solidification and curing kinetics and modulus build up...
- Simple solutions give good first estimates.