



WARSAW UNIVERSITY OF TECHNOLOGY
Faculty of Chemical and Process Engineering

Surface phenomena in separation of dispersed liquid contaminants using the coalescing filters

Andrzej Krasieński

e-mail: Andrzej.Krasinski@pw.edu.pl
phone: +48 22 234 64 93
address: Waryńskiego 1, 00-645 Warsaw

Presentation plan

Introduction

Principles of the coalescence filtration and applications

Surface and depth coalescence

Droplets behaviour on the fibers (SE, R-P instability)

Flow regimes, modelling of the proces

Experimental setup and some results

Coating of filters to obtain superhydrophobic and low-adhesion surface

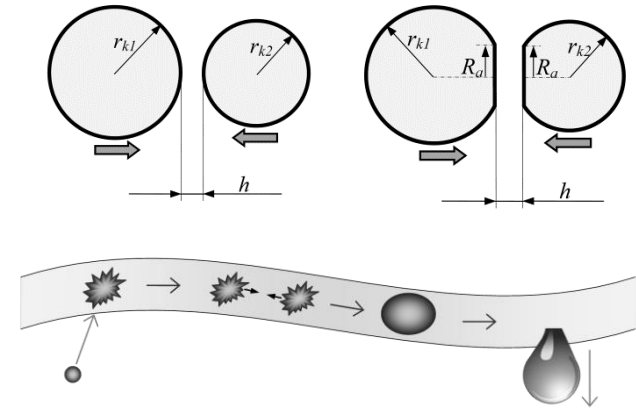
Modelling of the dynamic contact angles and droplet movement using CFD

Coalescence filters applications

Coalescence – process of merging droplets

For effective separation

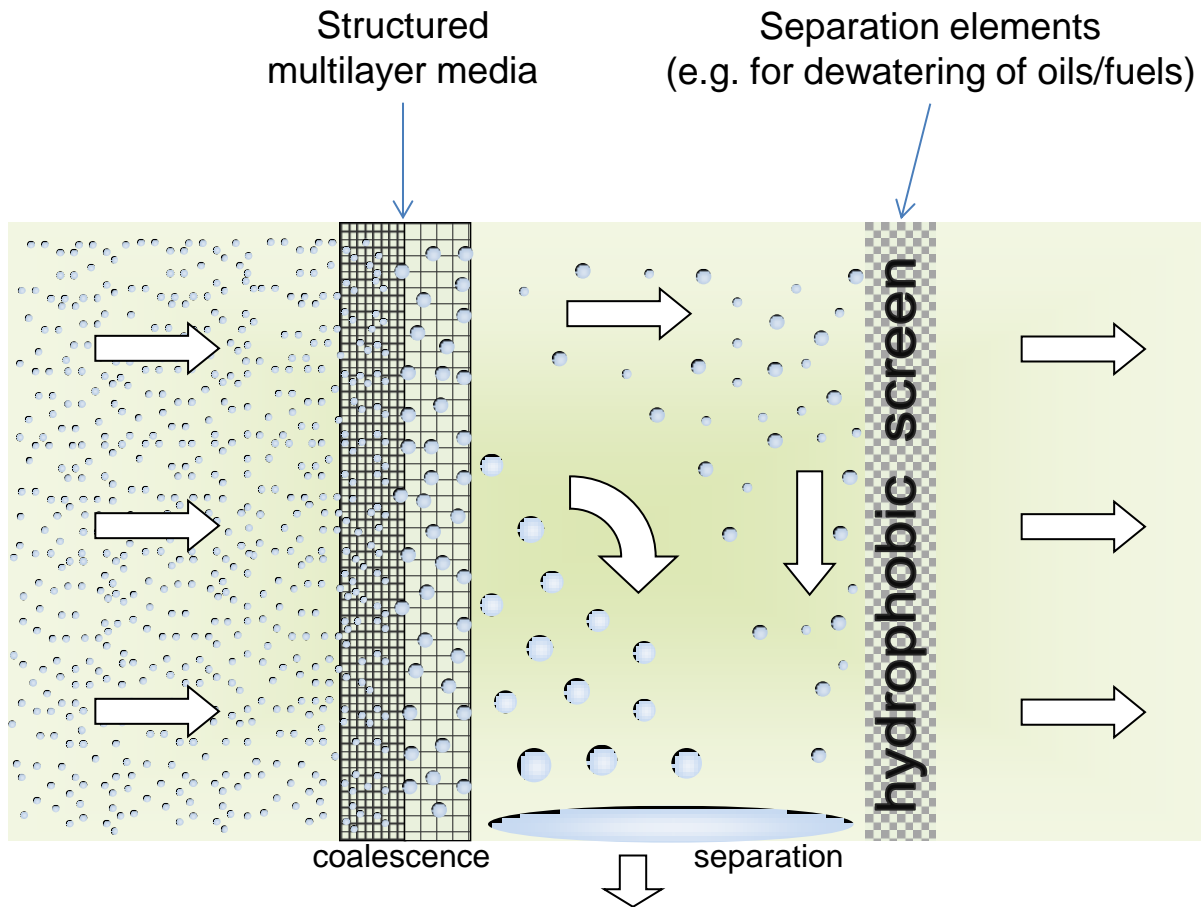
(by gravity, centrifugal force, membrane)
droplets must grow to a reasonable size



Four general categories of application:

- equipment protection (prevent corrosion, protect catalysts etc.),
- recovery of valuable products,
- obtaining high quality (purity) products – pollutants removal,
- meeting environmental discharge limits.

Principles of L/L coalescence filters separation



Principles of L/L coalescence filters separation

Coalescence media:

- deep bed process
- multilayer structures

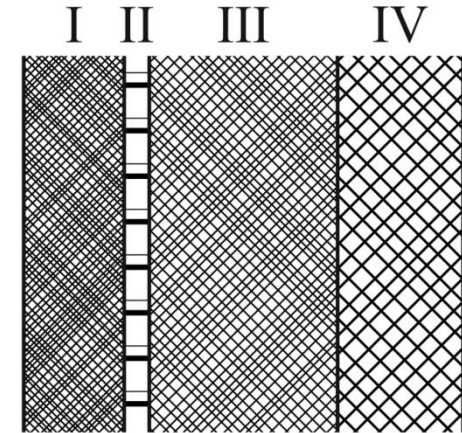
I-primary coalescence layer

II-support

III-intermediate (secondary coalescence) layer

IV-drain layer

flow →



Separation media:

- surface process

Automotive diesel filters (cellulose, fibreglass)

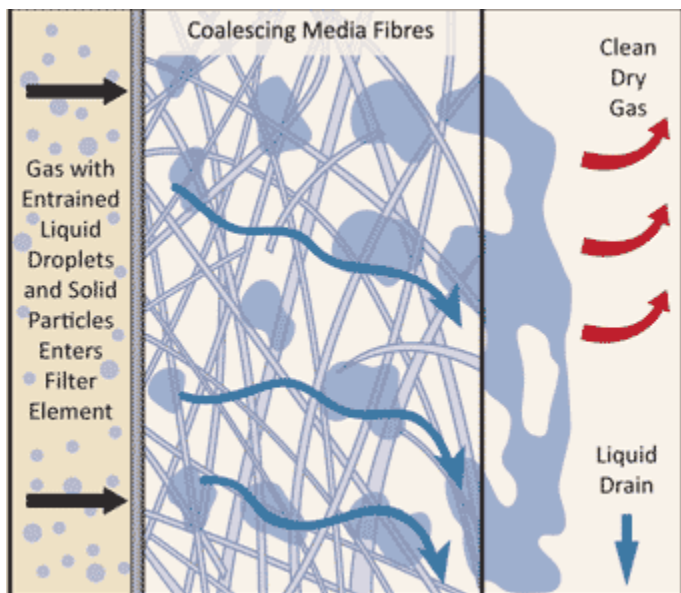
Separation elements

2nd stage for jet fuel (kerosene), diesel fuel
(Teflon coated SS mesh, hydrophobized nylon mesh,
hydrophobic fibrous cellulose)

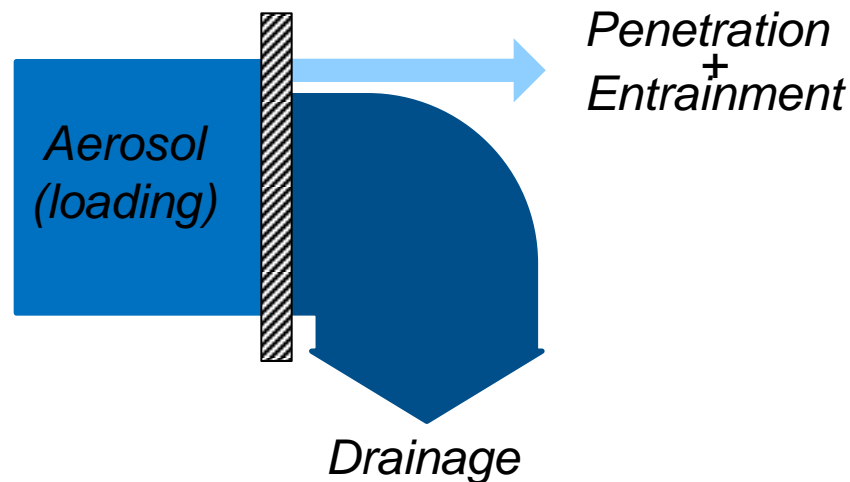
Principles of G/L coalescence filters separation

Difference comparing to L/L coalescers:

in properly operated filter liquid is usually drained in form of layer – on the outlet or internally (inside the porous structure)

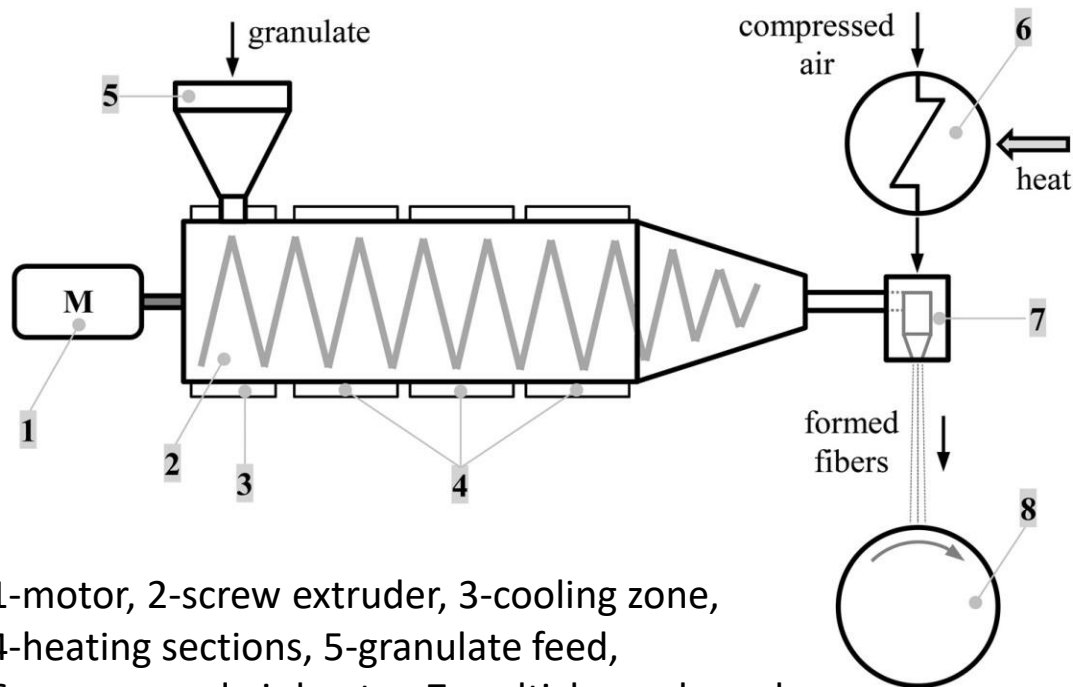


Picture taken from Kelburn Engineering website



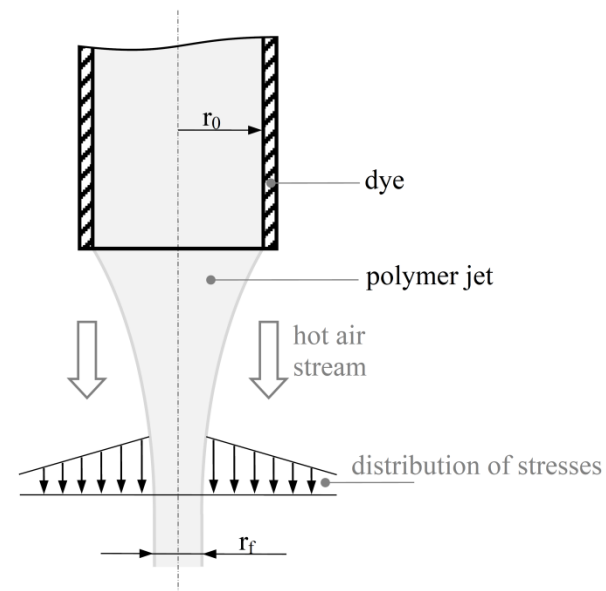
Coalescence filters manufacturing

Melt-blow technique (producing the polymer filter media)

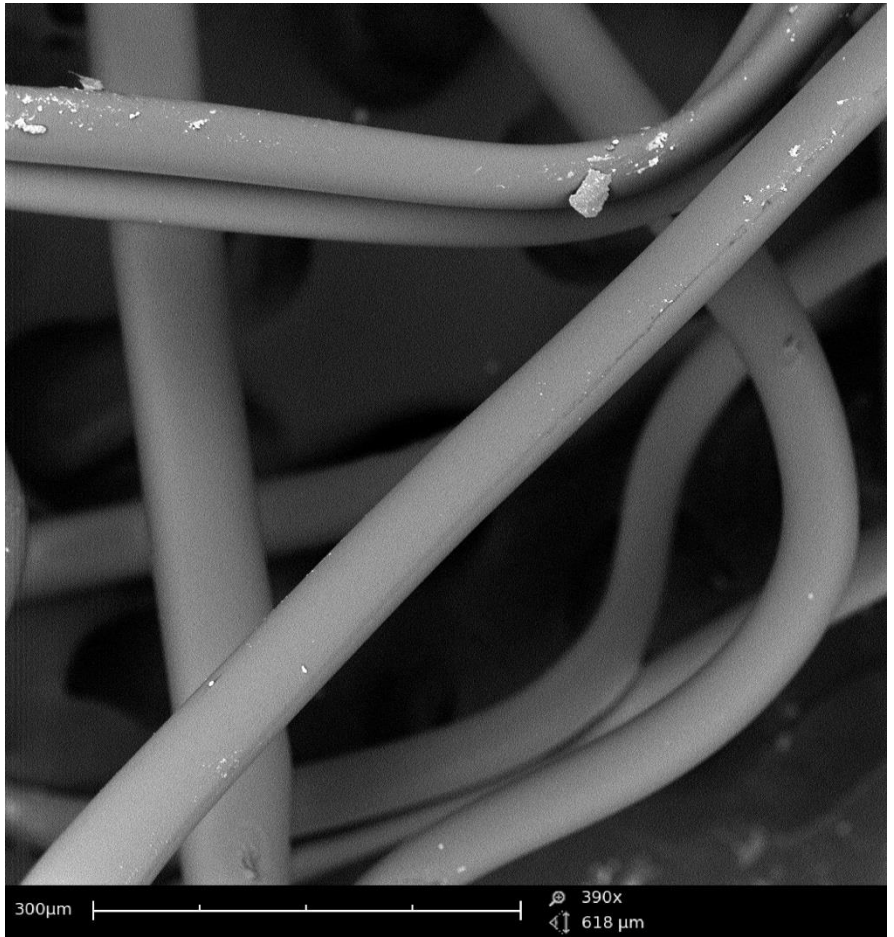
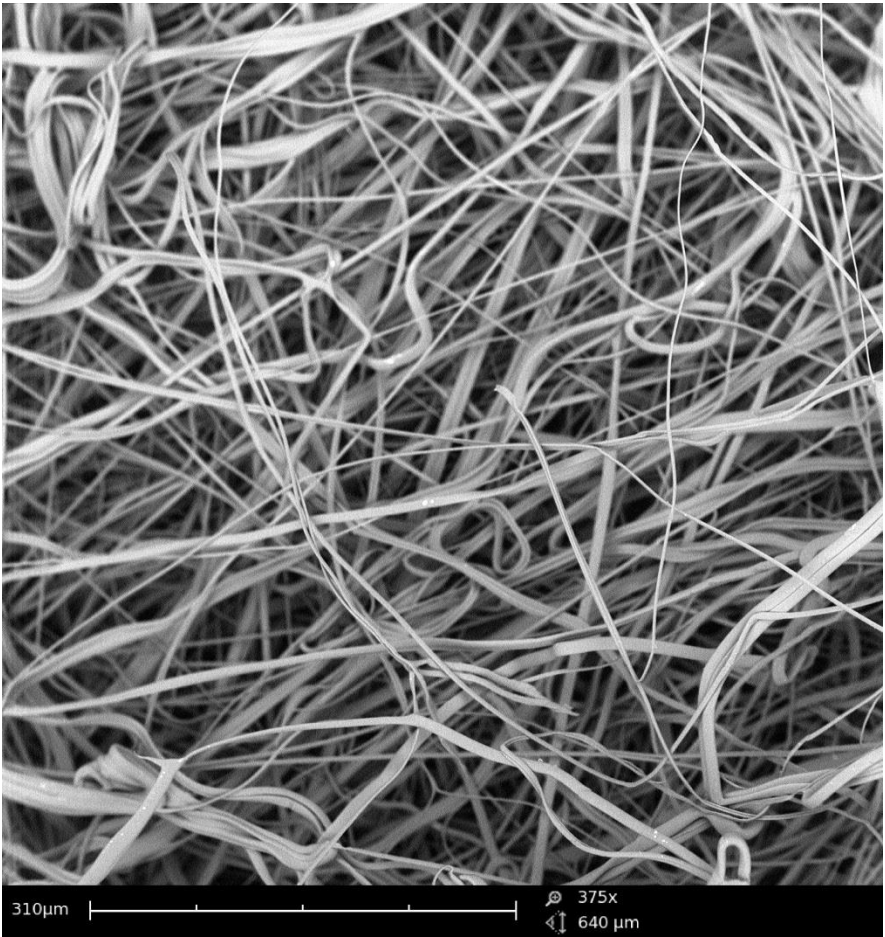


- 1-motor, 2-screw extruder, 3-cooling zone,
 4-heating sections, 5-granulate feed,
 6-compressed air heater, 7-multichannel nozzle,
 8-receiver drum

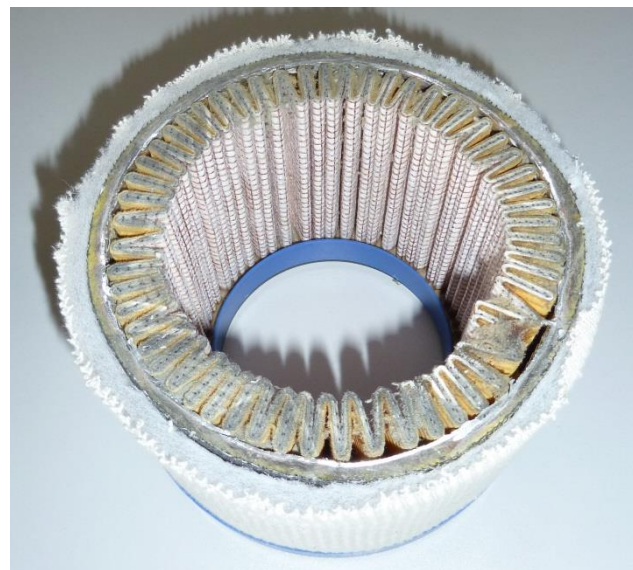
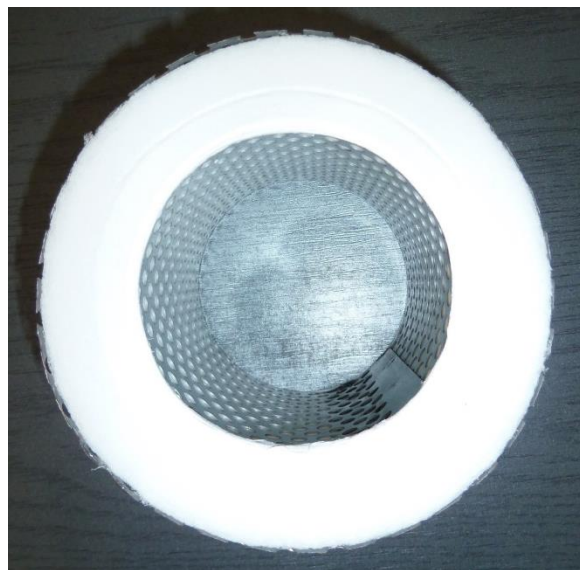
Fibre formation



Polymer fibrous media produced using the melt-blow technique

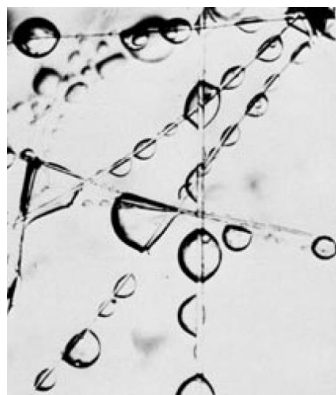
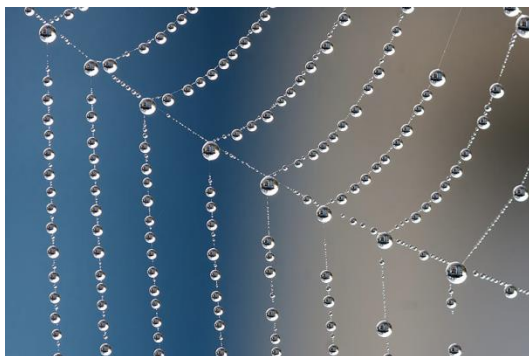


Coalescence elements

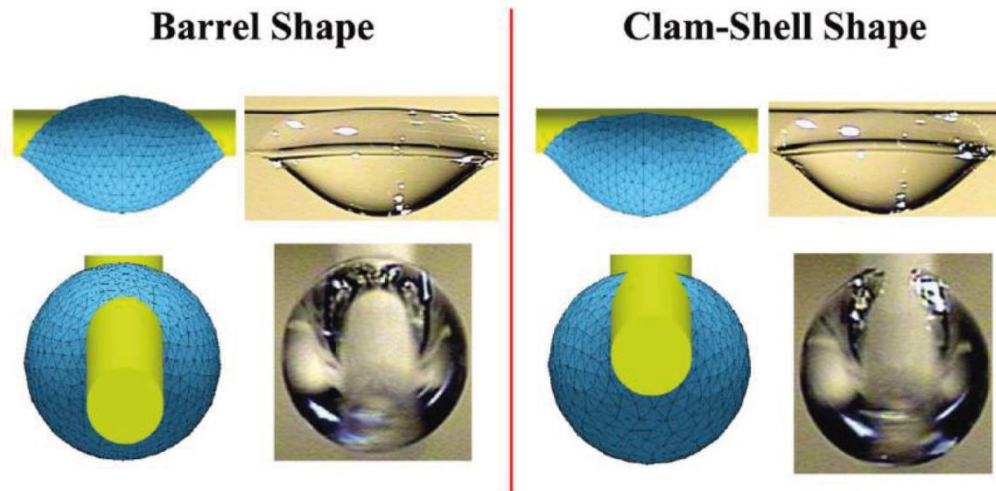


Droplets attached to fibres

A look into nature



Equilibrium geometric shape of a droplet on a fiber



Langmuir 2011, 27, 3685–3692

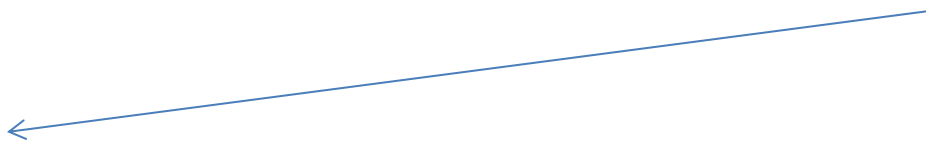
Surface Evolver (OA software) – minimizes the energy of a surface subject to constraints; the minimization by evolving the surface down the energy gradient

$$F = \gamma_{LG}A_{LG} + (\gamma_{SL} - \gamma_{SG})A_{SL} + \int \int \int_V (\rho g z) dV$$

Rayleigh-Plateau instability

A well-known Rayleigh-Plateau instability cause liquid jets or films to break into series or array of droplets.

e.g. melt-blow liquid on the fibres of coalescing filters



Even if the surface is perfectly wetted, the thickness of stable film is limited –
– the predominant wavelength of this instability is $\lambda = 2\pi\sqrt{2}r_f^*$

* Annu. Rev. Fluid Mech. 1999, 31, 347–384

The thickest possible film - when the wavelength is equivalent to the circumference of the liquid cylinder coating the fiber:

$$2\pi\sqrt{2}r_f = 2\pi(r_f + h_t)$$

$$h_t = r_f(\sqrt{2} - 1)$$

CFD
(OpenFOAM)

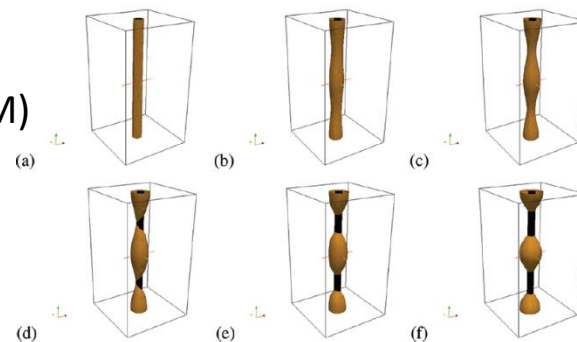


Figure 1. Series of images showing the simulation of the breakup of a liquid film under the influence of the Plateau–Rayleigh instability from (a) $t = 0$ to (f) $t = 0.00092$.

Modelling of the coalescence filtration

Operation:

1. collection of droplets onto fibres
2. merging on fibres to form large drops
3. detachment after reaching a certain size

Process is repeated within entire structure multiple times until reaching outlet

Inlet layers – capturing of small droplets

Outlet/drain layer – forming of large droplets (uniform in size)

Upstream the coalescence filter: dispersion of fine droplets

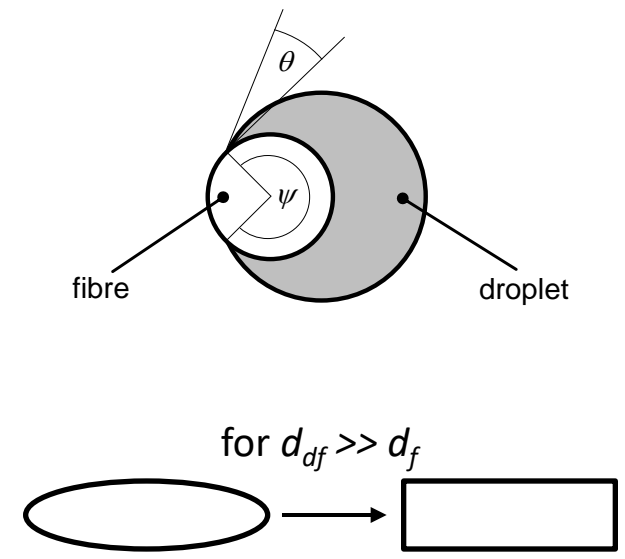
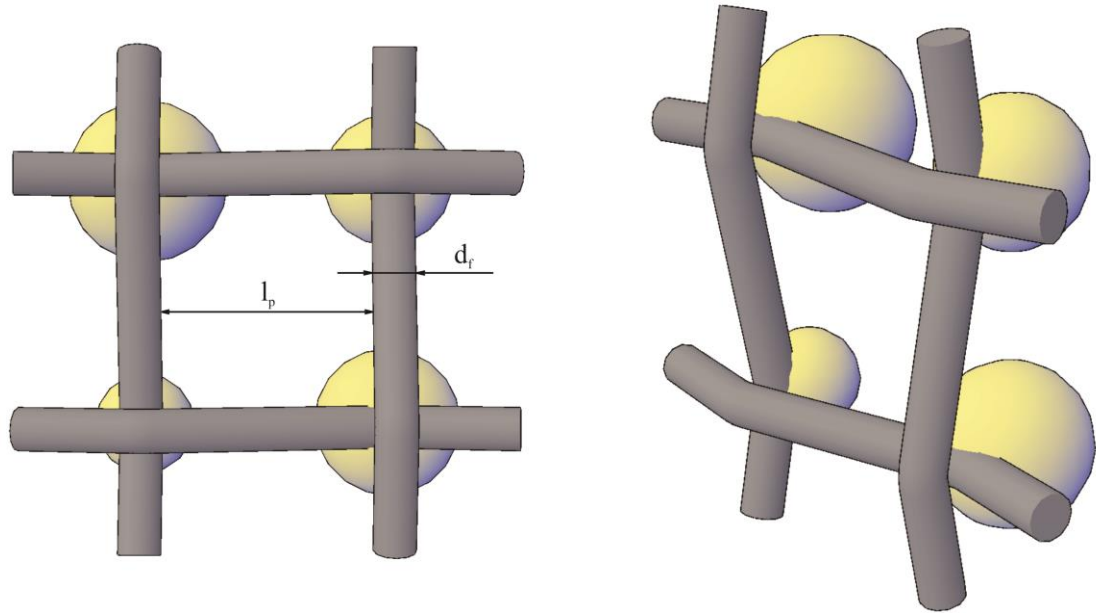
Downstream the coalescer: dispersion of very large droplets

Modelling of the coalescence filtration – model formulation

Computational cell

Assumptions:

- liquid accumulated in preferred locations (fibres intersections)
- immediate transport of all collected liquid to these locations
- no deformation



$$P = 2 d_{df} + d_f \psi$$

Modelling of the coalescence filtration – efficiency of deposition

In flow throughout porous media droplets can grow due to:

I. Fibre assisted coalescence:

- deposition directly onto fibres
- coalescence with droplets attached to fibres

II. Coalescence of free-flowing droplets due to viscous shear (shear-induced coalescence)

EFFICIENCY OF DROPLETS DEPOSITION

Fibre (cylindrical collector):

- Interception (Bürkholz, 1989)

(limiting trajectory concept)

$$y = \frac{d_f}{2} \frac{(1 + N_R) \ln(1 + N_R) - \frac{1}{2}(1 + N_R) + \frac{1}{2} \frac{1}{(1 + N_R)}}{Ku}$$

$$Ku = -0.5 \ln \alpha - 0.75 + \alpha - 0.25 \alpha^2$$

- Inertial impaction (Stechkina et al., 1969)

$$\eta_{in} = \frac{J Stk}{(2 Ku)^2} \quad J = (29.6 - 28 \alpha^{0.62}) N_R^2 - 27.5 N_R^{2.8} \quad Stk = \frac{d^2 \rho_p u C_C}{18 \mu d_f}$$

Droplet attached to the fibre (spherical collector):

- Interception (Lee and Liu, 1982)

$$\eta_R = \frac{1 - \alpha}{Ku} \frac{N_R^2}{1 + N_R}$$

- Inertial impaction (Langmuir and Blodgett, 1945)

$$\eta_{in} = \frac{Stk^2}{(Stk + 0.25)^2}$$

Modelling of the coalescence filtration – coalescence efficiency



Probability of deposition (when droplet approaches or collides the collector):

- for fibres = 100%

(observed efficiency of droplet deposition is equal to efficiency of species collection on a fibre due to considered mechanisms)

- for deposition on droplets attached to fibres – a formalism based on the film drainage model is applied (Chesters, 1991)

$$\eta_{coal} = \exp\left(-\frac{t_d}{t_c}\right)$$

t_d – drainage time (often referred as coalescence time)
 t_c – contact time

Relations for undeformed droplets were used in the model:

$$t_d = \frac{3 \pi \mu_c}{2 F} \left(\frac{d_{d1} d_{d2}}{d_{d1} + d_{d2}} \right)^2 \ln \left(\frac{h_i}{h_f} \right)$$

h_i – initial film thickness

h_f – final film thickness

F – interaction force (exerted by flow)

$$t_c = \frac{1}{\dot{\gamma}}$$

Modelling of the coalescence filtration – structure saturation

$$S = \frac{V_L}{V_0}$$

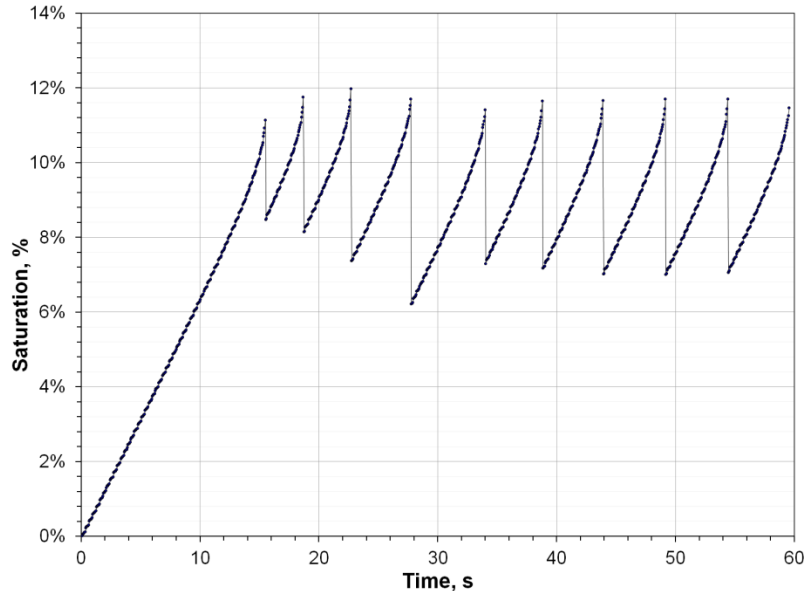
volume occupied by dispersed phase liquid
total volume of voids

For thin and uniform media
it can be estimated based on
the pressure drop:

$$\frac{\Delta p_1}{L} = \frac{36 \cdot u \cdot \mu_c \cdot K_1 \cdot (1 - \varepsilon_1)^2}{d_F^2 \cdot \varepsilon_1^3}$$

$$\frac{\Delta p_2}{L} = \frac{36 \cdot u \cdot \mu_c \cdot K_2 \cdot (1 - \varepsilon_2)^2}{d_F^2 \cdot \varepsilon_2^3}$$

$$S = 1 - \frac{\varepsilon_2}{\varepsilon_1}$$



Time-dependent
saturation profile:
 $\alpha=0.001$,
 $d_d=5\mu\text{m}$,
 $d_f=10\mu\text{m}$,
 $l_p=100\mu\text{m}$,
 $u=10\text{mm/s}$,
 $c=1\% \text{vol.}$,
 $\theta=30^\circ$.

	u	1 mm/s	5 mm/s
$\theta = 30^\circ$	S	10.3 %	9.97 %
	A _F	0.6-30.3 %	3.2-31.7 %
$\theta = 60^\circ$	S	7.32 %	6.87 %
	A _F	4.2-40.4 %	11.5-42.7 %

	10 mm/s	50 mm/s	100 mm/s
	9.13 %	8.12 %	4.19 %
	6.5-34.1 %	16.5-38.4 %	36.5-46.9 %
	6.02 %	5.23 %	2.11 %
	18.6-45.1 %	32.7-50.9 %	47.5-59.9 %

Modelling of the coalescence filtration – detachment of droplets

Two independent criteria for droplets detachment from fibres are considered:

1. stable size of a droplet in local hydrodynamic conditions
2. balance of adhesion and drag forces

Ad. 1.

Walstra 1993: $We_{crit} = \frac{\mu_c \dot{\gamma} d_{max}}{\sigma} = 2$

For the plug flow in fibrous bed: ~~$\dot{\gamma} = \frac{4u(1-\varepsilon)}{d_f \varepsilon^2}$~~

Shear rate in saturated bed: $\dot{\gamma} = \frac{2u_{local}}{l_{pore}^{eq}}$

Ad. 2.

$$F_D = \frac{\frac{15}{2} \pi d_{df} u_{local} \mu_c \left[1 + \frac{2}{3} \frac{\mu_c}{\mu_d} \right]}{1 + \frac{\mu_c}{\mu_d}}$$

vs. $F_A = \sigma \cos(\theta) P$

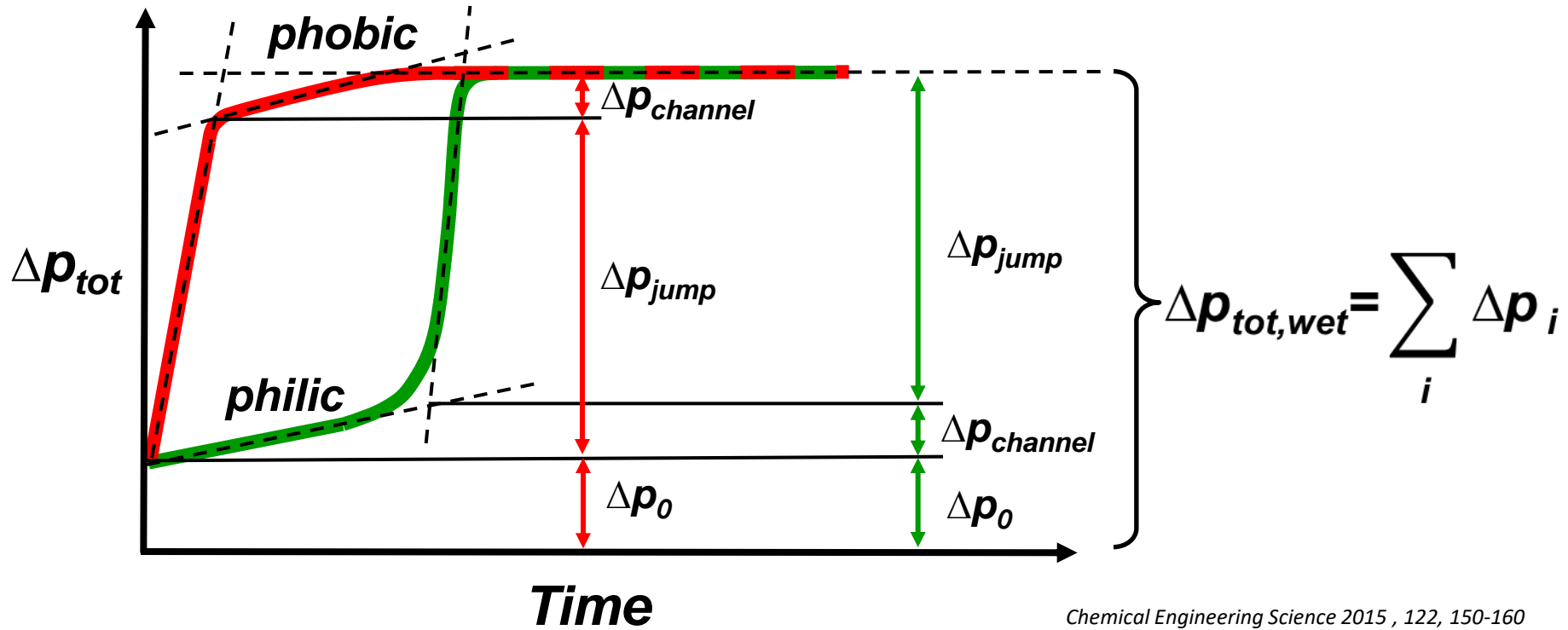
	d_f	5 μm	10 μm	25 μm	50 μm	100 μm
θ 30°		52.7 _S	108 _S	263 _S	496 _S	1063 _S
θ 75°		73.2 _{D-A}	146 _{D-A}	366 _{D-A}	729 _{D-A}	1457 _{D-A}

Mean size of detached droplets (in μm) as a function of fibre diameter and contact angle;

calculations for superficial velocity 10 mm/s, packing factor 0.001, $l_p = 10 d_f$;

Superscripts indicate the detachment criterion: S – stable size of droplets, D-A – drag to adhesion force ratio.

Film and channel model

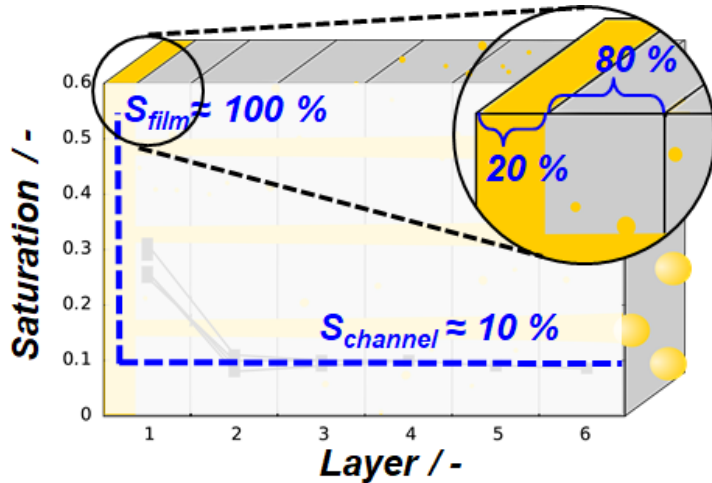


Chemical Engineering Science 2015 , 122, 150-160

Δp_{jump} : associated with film (capillary phenomenon depending only on media properties)

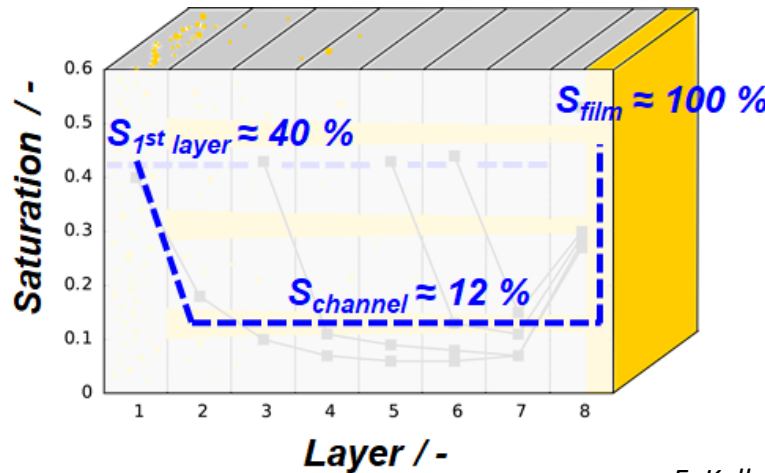
$\Delta p_{channel}$: associated with oil channels ("flow phenomenon" depending on viscosity, droplets load, face velocity,...)

Film and channel model – saturation profiles



Sections of const. saturation in phobic media:

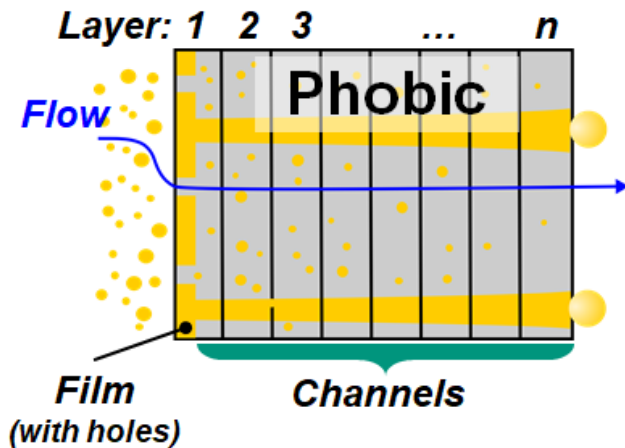
- Film (thickness $f \approx 20\%$ of 1st layer)
- Channels



Sections of const. saturation in philic media:

- 1st Layer
- Channels
- Film ($f \approx 20\%$ of last layer)

Film and channel model – efficiency of multilayer filter



Penetration (1 – efficiency) depends on:

- properties of fibrous structure
- saturation

For the sandwich of filtration layers (the same media) the total penetration:

$$P = P_1 \cdot P_2 \cdot P_3 \dots P_n$$

P_i 's: "sectional penetrations"

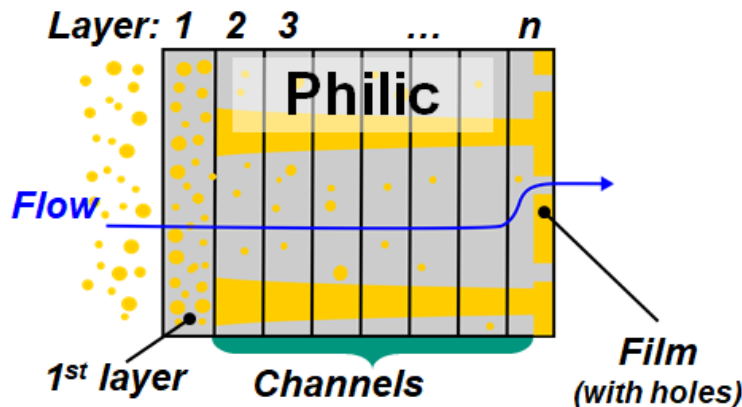
→ Phobic sandwich of n layers:

$$P = P_{\text{film}} (P_{\text{channel}})^{n-f}$$

→ Philic sandwich of n layers:

$$P = P_{1^{\text{st}} \text{ layer}} (P_{\text{channel}})^{n-1-f} P_{\text{film}}$$

where f is film thickness in % of single layer



Modification of the filter media

Methods (for polymer media):

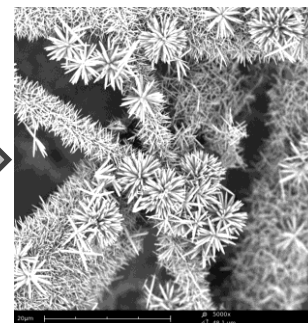
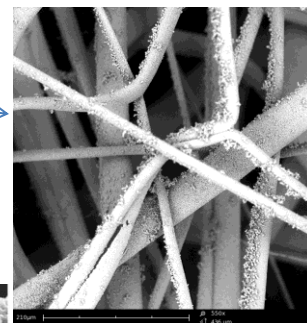
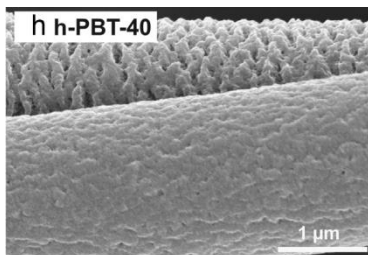
- primary, i.e. at the stage of fibre formation
- secondary, i.e. coating of fibrous structure

Aim:

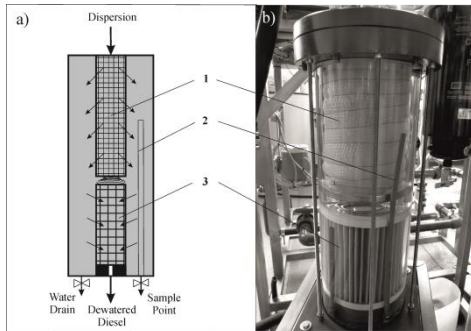
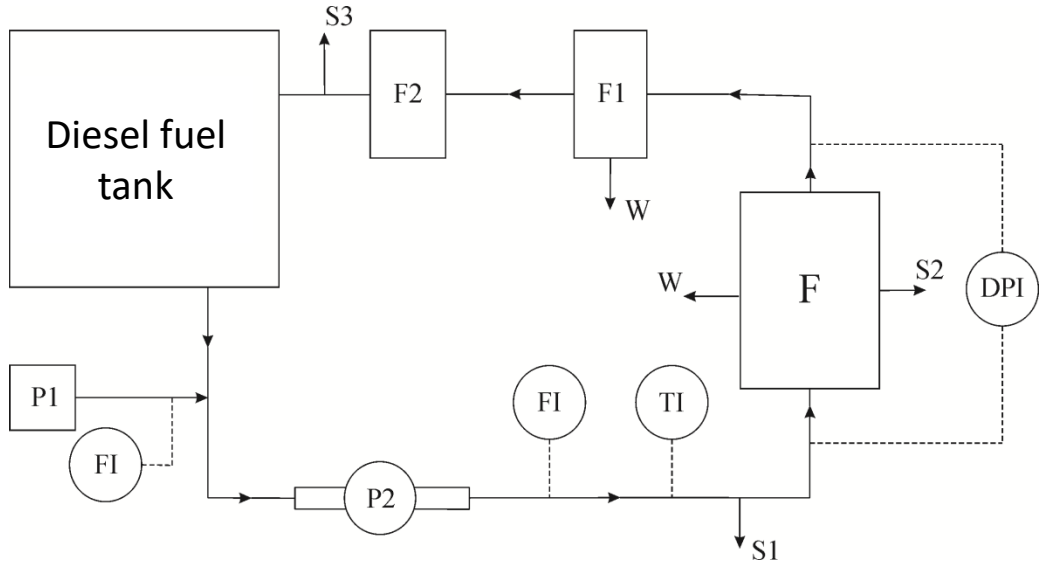
- change of wettability (obtain desired affinity with a liquid)
- antimicrobial properties (bacteria, algae, molds)
- catalytic/photocatalytic properties
- sorption (organics, metal ions)
- improve electric conductivity

Examples:

- Chemical additives to polymer matrix – prepare masterbatch or twin screw extruder (e.g. Halar®1400LC oraz FluoroLink® added to PP)
- ZnO needles deposited on the fibres (CBD) →
- High-voltage electric field treatment of the molten polymer (during the production)
- Low temperature plasma treatment
- Hydrolysis of polyester fibre surface →

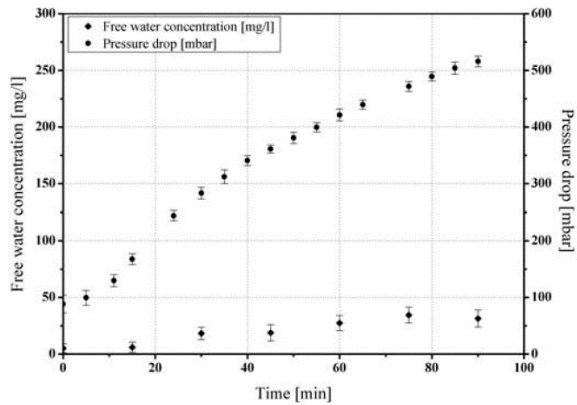


Experimental – test rig



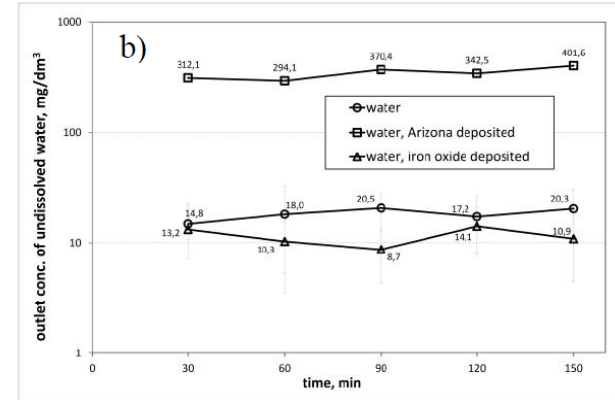
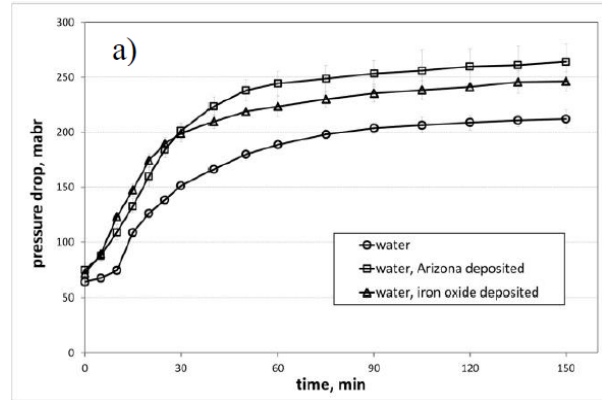
Experimental – some results

Perfectly water wetted fibreglass media

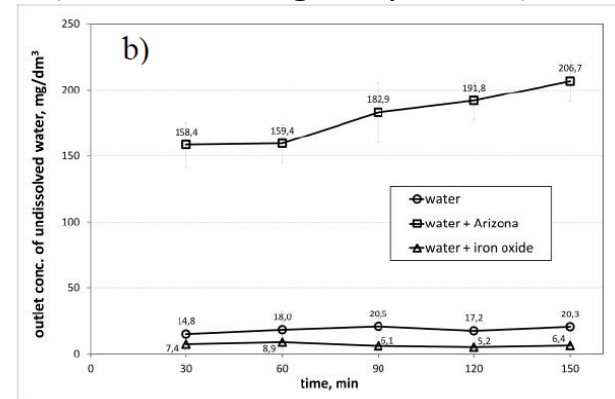
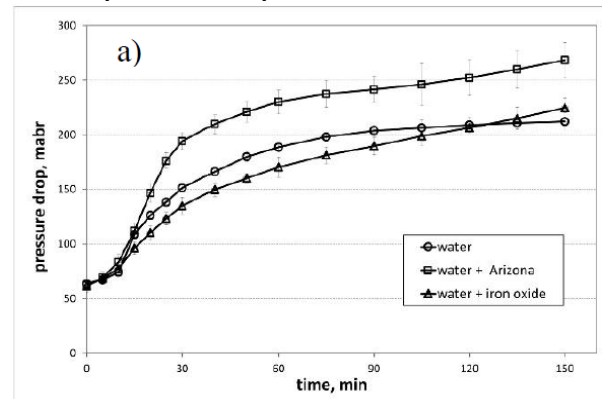


Effect of solids

particles deposited in fibrous media before experiment



particles present in the emulsion (filtered during the process)

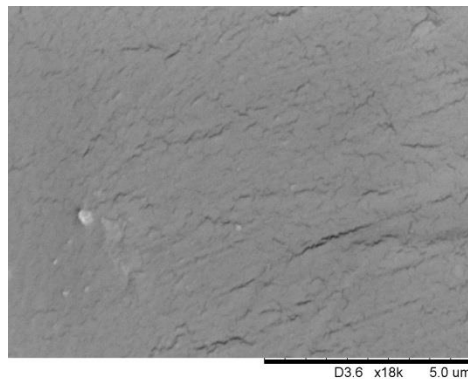


Experimental – some results

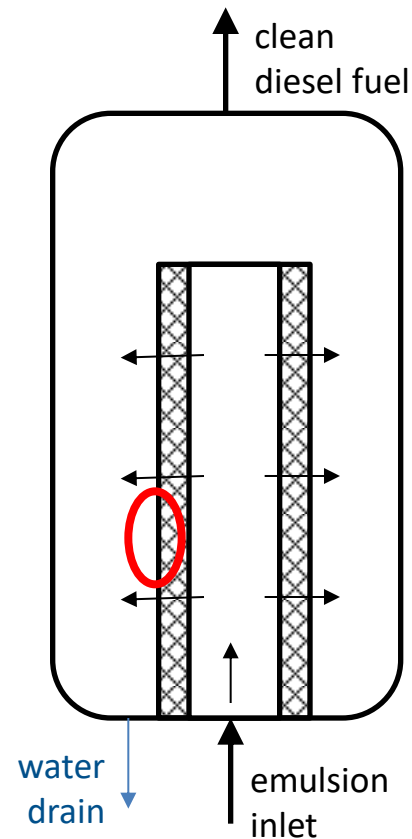
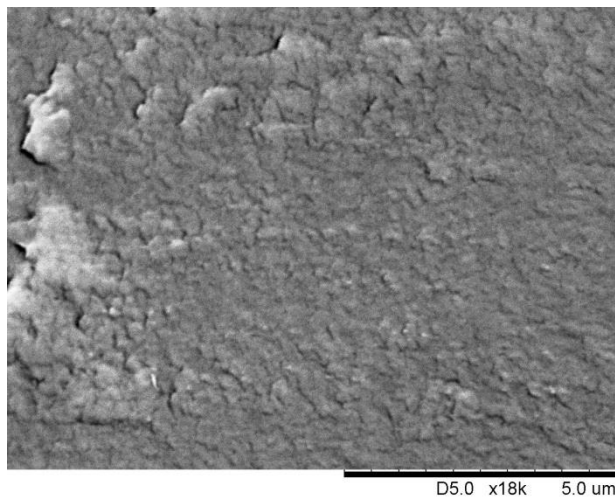
Effect of surface treatment (PBT native and modified with Ar/O₂ plasma)

SEM images of the PBT fibers surface:

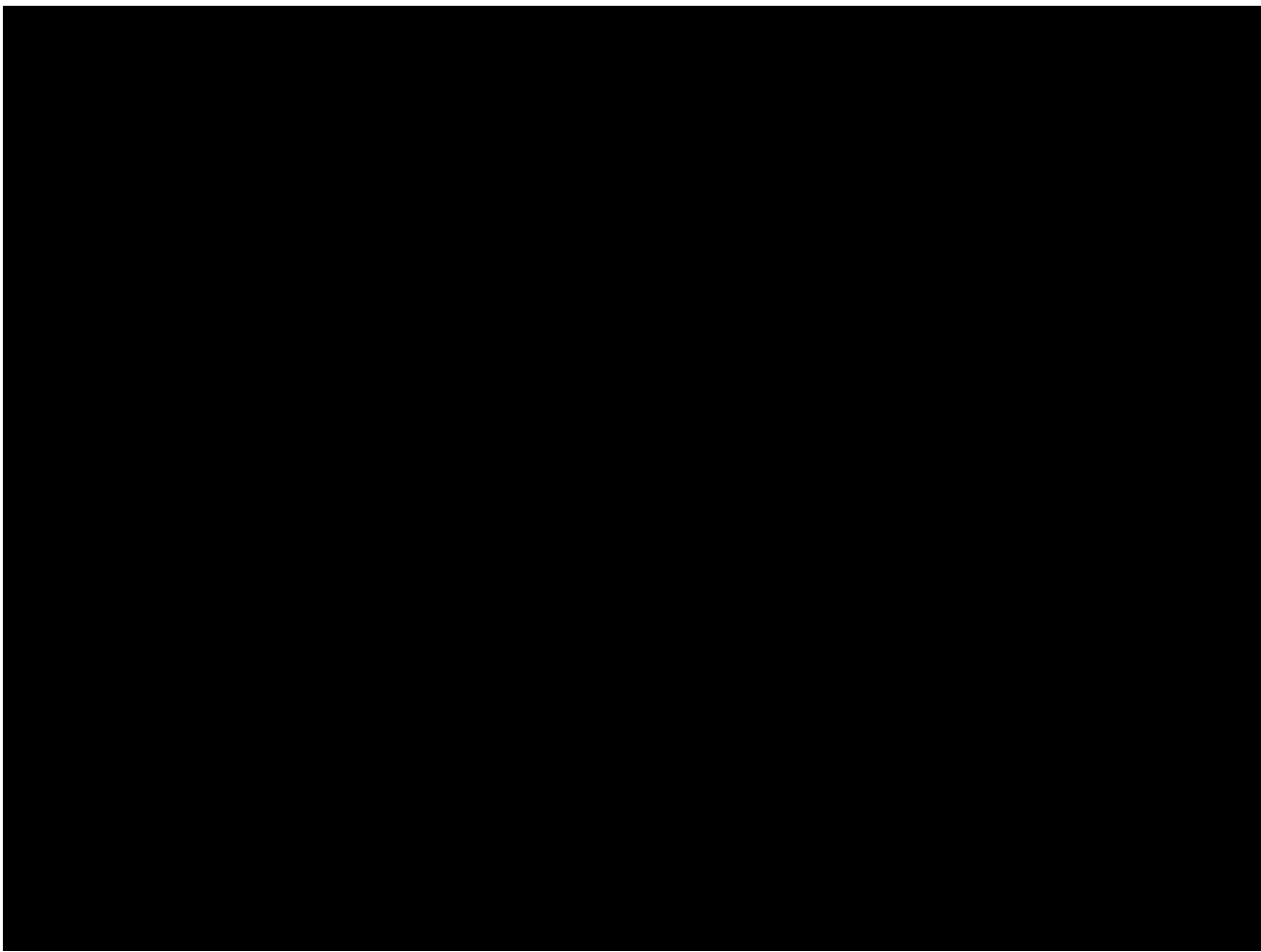
native (untreated)



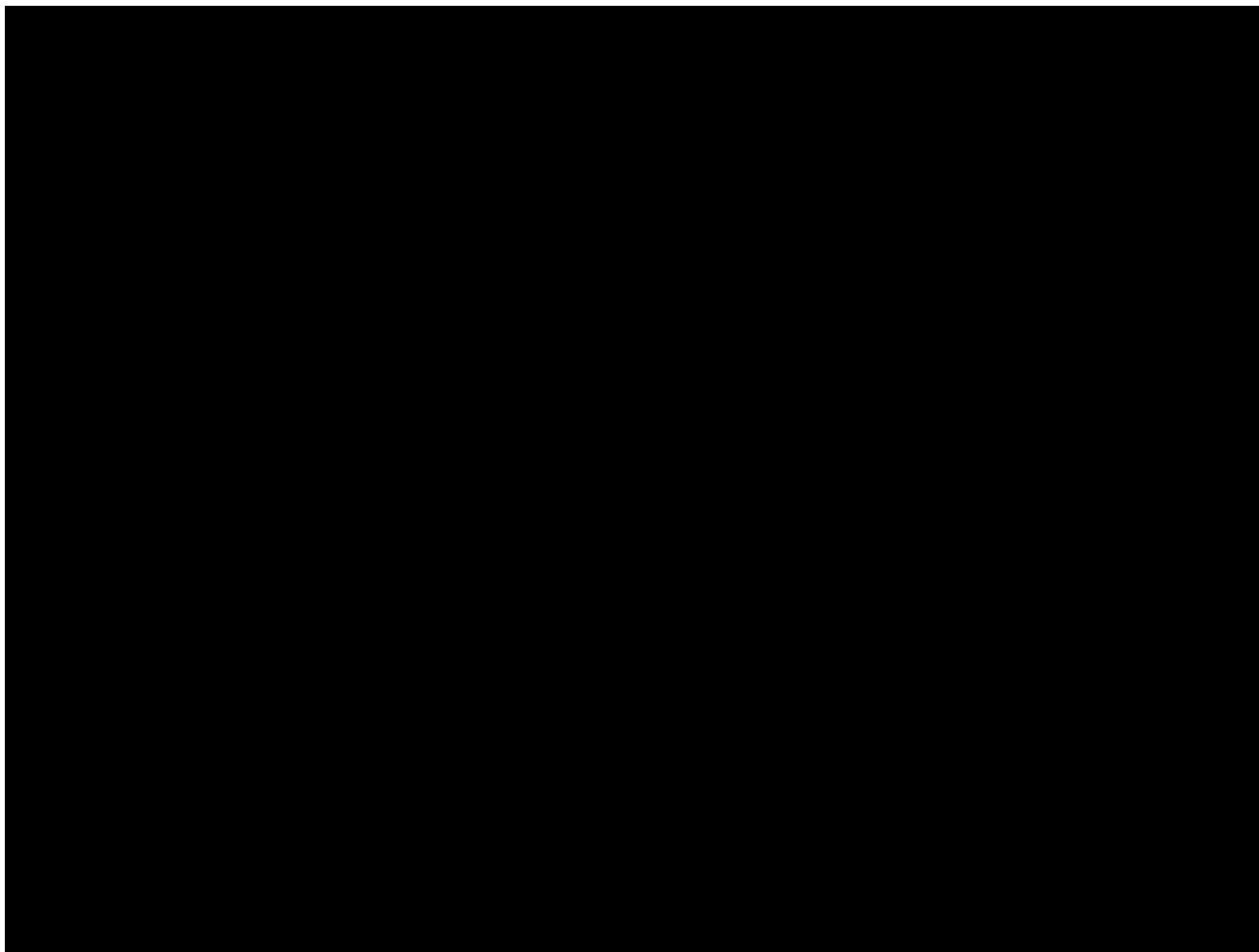
after plasma process in the O₂/Ar = 40/60 vol. gaseous mixture (treat. time 1.5 min, discharge power 50 W, voltage -550V, abs. pressure 1 mbar)



Experimental – untreated PBT



Experimental – plasma modified PBT



Experimental – drain layer design

Water deoiling (separation of O/W emulsion) – PP versus PA



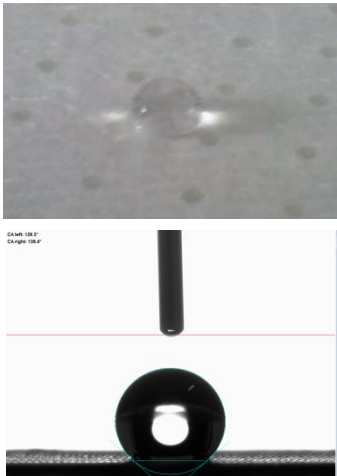
Surface coalescence

COALESCER (I) 1. **Increase size of droplet size upstream the separation**

SEPARATOR (II) 2. **Separate the droplets by gravity or using te separation filter (capillary forces):**

- apparent pore size, packing density, fibers size (specific surface area)
- phobicity of filter media with the dispersed phase (no wettability: superhydrophobicity for dewatering applications)
- improvement of self-cleaning abilities of separators (e.g. low adhesion to the surface + hydrodynamics)
- very low face velocity

$$dP < \text{„capillary pressure”}$$



Specific process: water removal from diesel fuel

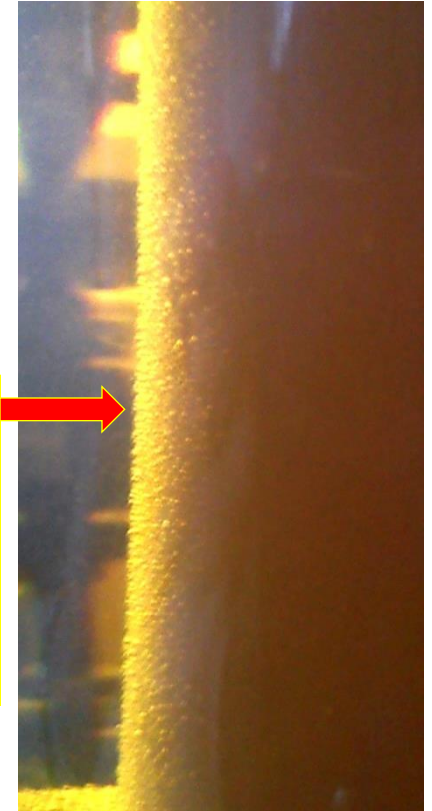
Origin of problems in recent years:

- ULSD
- FAME
- additives
- high quality of fuel necessary for CR injection systems

These led to:

- decrease of IFT value
(clearly less than e.g. 15-19 mN/m as per SAE J1488)
- increase of viscosity
(biodiesel blends – also vulnerability to bacteria growth)
- increase of water solubility

hydrophobic
filtration
surface
„coated”
by water
droplets



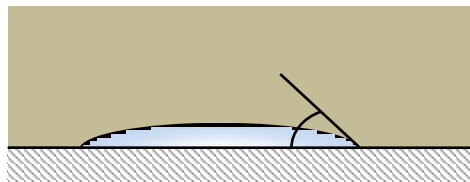
**Hindered coalescence of water droplets, no drainage –
– accumulation of water on surface, above certain dP filter breakthrough**

Wettability – surface properties determination

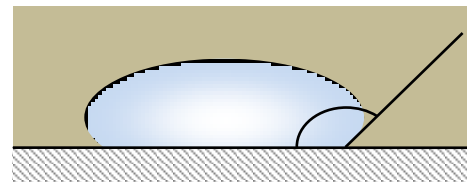
Static contact angle



High surface energy (and σ_{crit})
Good wetting (0° contact angle)

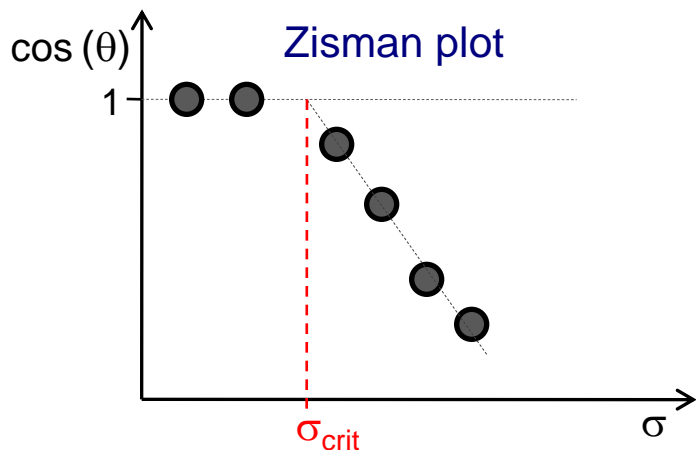


Wetting
($0^\circ < \text{contact angle} < 90^\circ$)



Low surface energy (and σ_{crit})
No wetting (contact angle $> 90^\circ$)

Critical surface tension



Free surface energy can be determined using one of following methods:

- geometric mean (based on *Fowkes eqn*)
- harmonic mean (method derived by *Wu*)
- acid-base method (*van Oss and Good*)

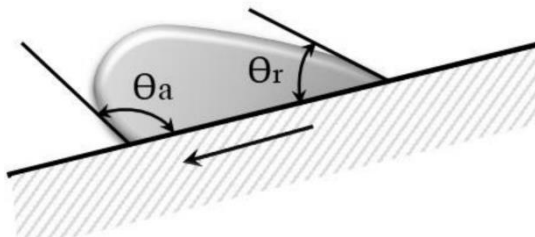
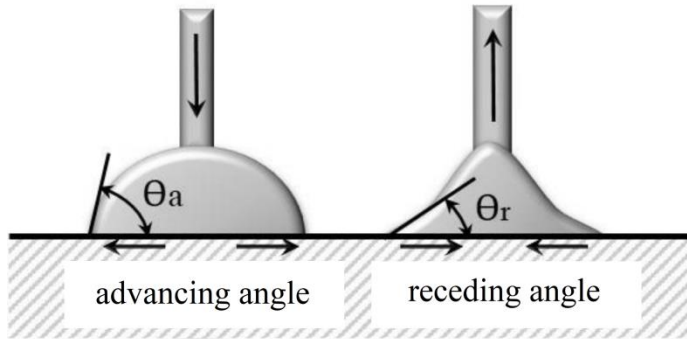
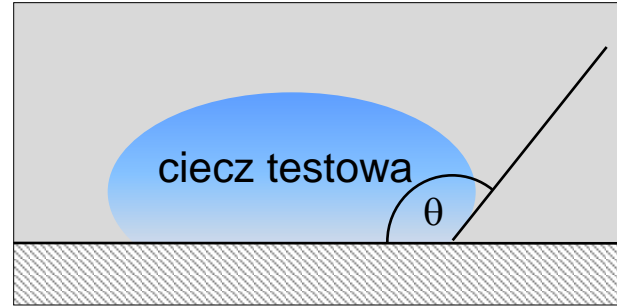
and few other methods

Serie of test liquid – homologs, e.g. n-alkanes, alcohols

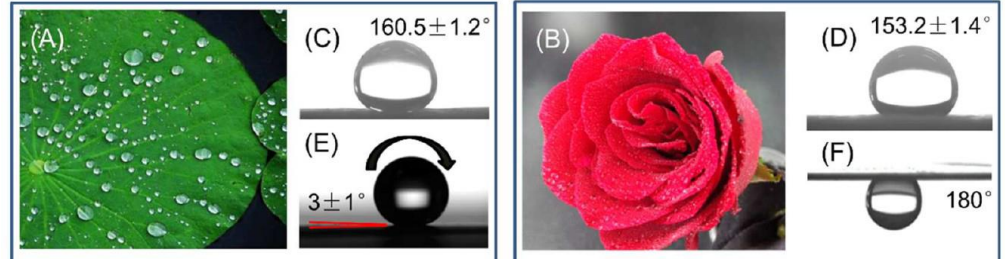
Wettability – determination of surface properties

For water separators applied for dewatering of diesel fuel:

- hydrophobic surface
- (superhydrophobic $\theta > 150^\circ$)



A look into nature – superhydrophobic surfaces



Lotus leaf – low adhesion Rose petal – high adhesion

Ind. Eng. Chem. Res. 2017, 56, 907–919

$$\rho_k g V_k \sin(\alpha) = 2 R_{zw} k$$

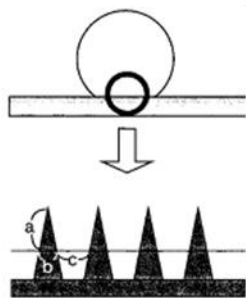
$$k = \sigma [\cos(\theta_r) - \cos(\theta_a)]$$

Modification of filter media applied for diesel fuel dewatering

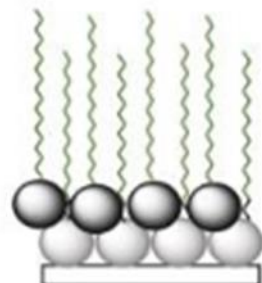
HIGHER
HYDRPHOBICITY

Surface morphology
(roughness)

Chemical
composition



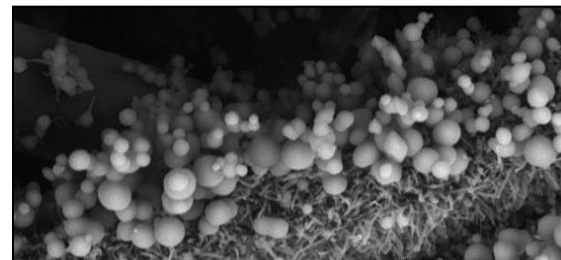
(Miwa et al., 2000)



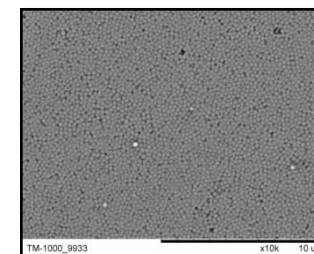
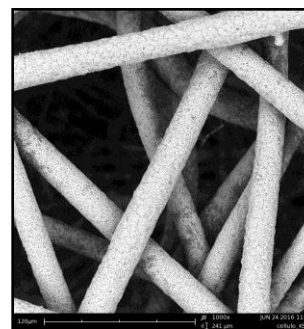
(Xue et al., 2011)

Chemical compounds
with low surface
energy,
e.g.: fluorocarbons,
silicones, silanes.

Wenzel and Cassie-Baxter models –
effect of the morphology



Fiber
coated
with
aerogel
particles



Fibers coated with
silica particles

HYDROPHOBIZATION BY DIP-COATING METHOD

CA: $156.4^\circ \pm 4.0^\circ$

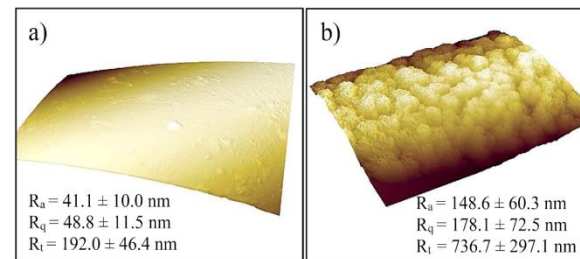
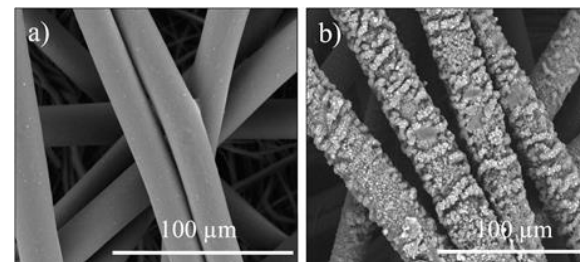
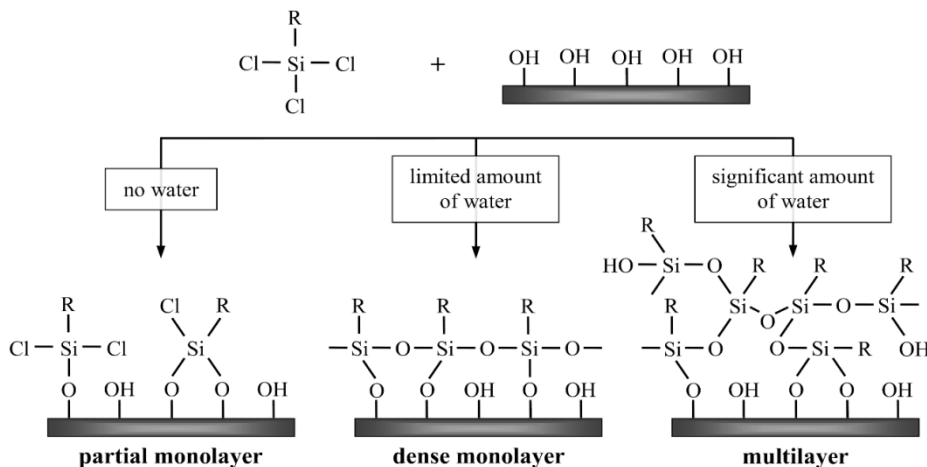
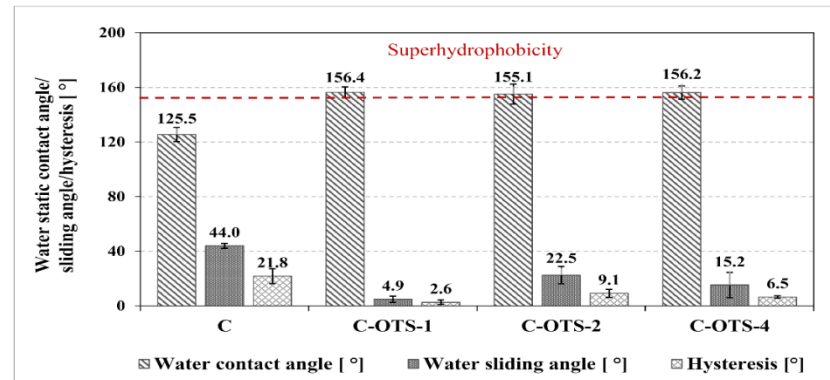
$$\begin{array}{c}
 \text{OH} \quad \text{OH} \quad \text{OH} \quad \text{OH} \quad \text{OH} \\
 | \quad | \quad | \quad | \quad | \\
 \text{---} \text{Si} \text{---} \\
 \text{Polyester fibers}
 \end{array}
 +
 \begin{array}{c}
 \text{R} \\
 | \\
 \text{Cl} \text{---} \text{Si} \text{---} \text{Cl} \\
 | \\
 \text{Cl} \\
 \text{Octadecyl-} \\
 \text{trichlorosilane} \\
 \text{(OTS)}
 \end{array}
 =
 \begin{array}{c}
 \text{R} \quad \text{R} \quad \text{R} \quad \text{R} \\
 | \quad | \quad | \quad | \\
 \text{HO} \text{---} \text{Si} \text{---} \text{O} \quad \text{Si} \text{---} \text{O} \quad \text{Si} \text{---} \text{O} \quad \text{Si} \text{---} \text{O} \quad \text{Si} \text{---} \text{R} \\
 | \quad | \quad | \quad | \\
 \text{---} \text{Si} \text{---} \text{O} \quad \text{O} \text{---} \text{Si} \text{---} \text{R} \\
 | \quad | \quad | \quad | \\
 \text{O} \quad \text{OH} \quad \text{O} \quad \text{OH} \quad \text{O} \quad \text{OH} \\
 \text{Multilayer OTS coating}
 \end{array}$$

Some experimental results for OTS-coated separation media

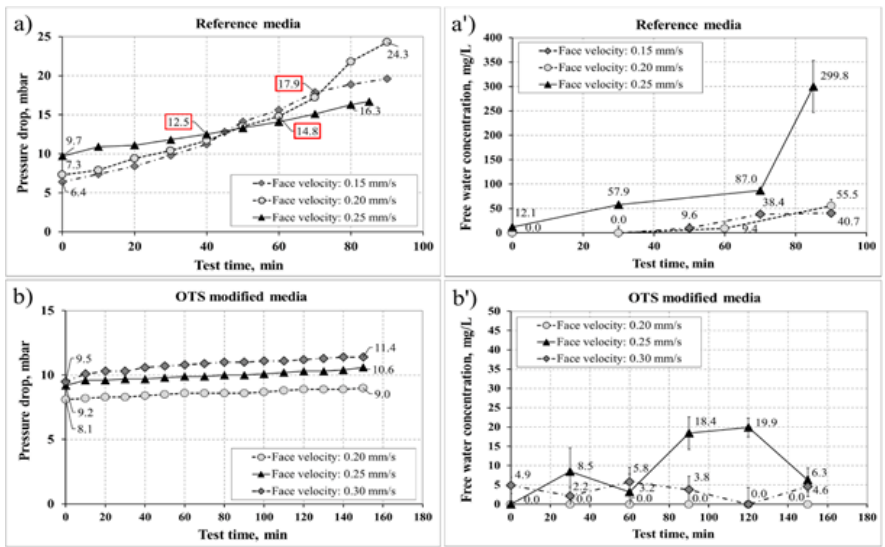


350 H/SM50 (Ahlstrom)
 2 polyester prefiltration layers on the cellulose:
 1) spun-bond PET
 2) melt-blown PBT

Top view of the pleated separator

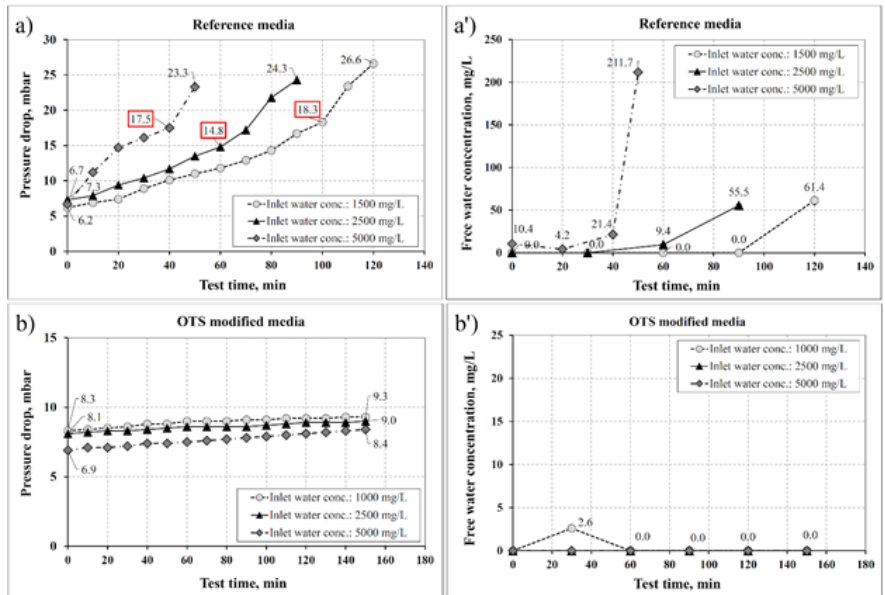


Some experimental results for OTS-coated separation media



The effect of the face velocity on the pressure drop and free water concentration on the outlet

The effect of the inlet water concentration



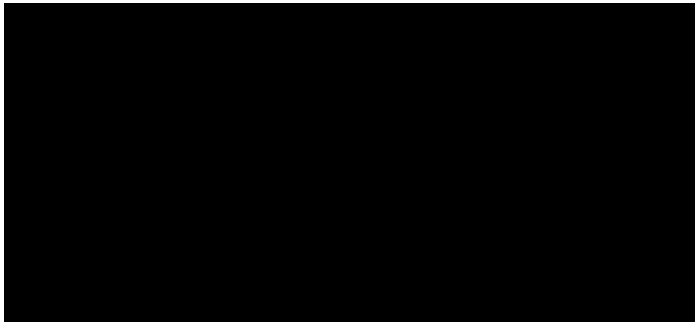
Modelling the dynamic contact angles and droplet movement using CFD

Ongoing work of PhD student – Patrycja Jachimczyk (Wierzba)

Measurements
of CAs and roll-off angle
for surface modified media

Development of UDF code, which will include:

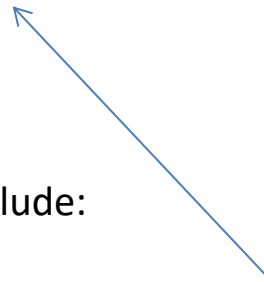
- defining CAs at the interface
- pinning (i.e. immobilizing) the droplet
- extracting the CAa at the droplet symmetry plane
- calculating the total force acting upon the droplet



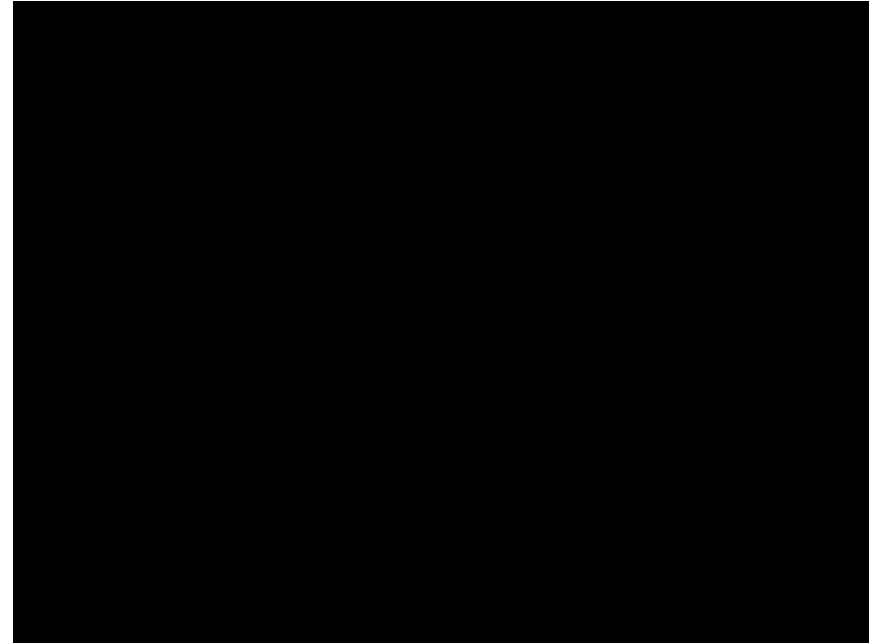
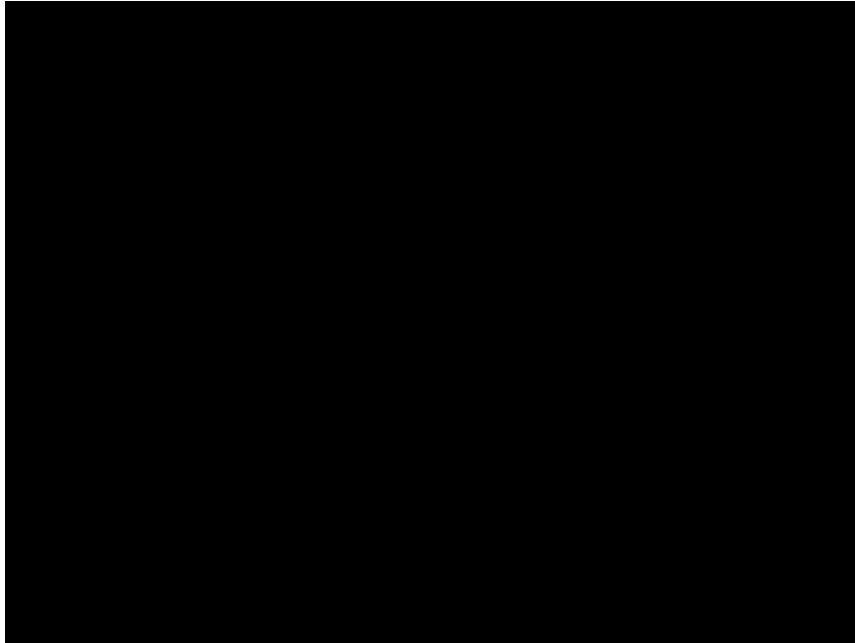
Introducing a new relation
for dynamic contact angles
depending on force acting
upon the „pinned” droplet

Comparing the force
(gravity component or drag)
at the onset of motion

Analysis of the droplets mobility
depending on surface properties
of filter media, droplet size and
hydrodynamics at the surface



Modelling the dynamic contact angles and droplet movement using CFD



Thank you for your attention