Flow in porous materials: a of tale X-rays, minimal surfaces and wettability

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An introduction to Imperial College London





Research-led university in the heart of London specialising in science, engineering, medicine and business

- Top 10 university globally
- Largest research income in UK
- Largest number of science, engineering and medical students
- Honours
 - 14 Nobel prizes
 - 57 Fellows of Royal Society
 - 91 Fellows of the Royal Academy of Engineering
 - 71 Fellows of Academy of Medical Science

Our mission statement



Imperial College London's mission is to achieve enduring excellence in research and education in science, engineering, medicine and business for the benefit of society.

Our people

Students

17,568 in total (2018-2019)

- 10,054 undergraduate
- 3,864 taught postgraduate
- 3,650 research postgraduate
- Students from 130+ countries

Staff

- 4,000 academic and research staff
- 4,000 support staff

Alumni

Over 190,000 alumni



Our students



Department of Earth Science & Engineering



Staff & Students

Staff

45 academic80 research staff30 support staff

Students

300 – undergraduate200 – taught postgraduate150 – research postgraduate

Total

Around 800 individuals



Undergraduate Teaching

Undergraduate courses:

- Geology
- Geophysics
- Planetary Science

Graduate with either BSc or MSci degrees

Top ranked in the Guardian and 2nd in THES and Complete University Guide





Postgraduate Teaching

Postgraduate taught courses:

- Petroleum Engineering
- Petroleum Geoscience
 Approximately 80 students
 Large international group
- Mineral and Energy Finance
 with Business School
 30 students
- Applied Computational Science & Engineering

120 students





Research Sections

Earth & Planets: E&P (17 staff)



Tidal environments & facies analysis

Natural Resources Geoscience and Engineering NRGE (17 staff)



Modelling tidal/heterolithic reservoirs (tidal cross-bedding & heterolithic facies)

Computational Geoscience and Engineering (15 staff)



IC Ocean Modelling (reconstruction of paleo-tides in South China Sea)

- Three research sections
- Multi-disciplinary research
- Fundamental science provides the insight to solve applied problems
- Engineering solutions provide the technology to solve problems and, simultaneously, improve understanding in fundamental science

Natural Resources Geoscience & Engineering







Reservoir Characterization & Modelling





Pore Scale Modelling



Rock Mechanics & Fractured ReservoirsGranular SystemsReservoir Geophysics-FWIWell Test Analysis

Carbon Storage

Carbonate Research



Basins Research & Geodynamics

Petroleum Research Applications:

- Basin & play analysis
- Reservoir Geophysics-FWI
- Reservoir characterisation & modelling
- 'Next generation' reservoir simulation models
- IOR/EOR applications in conventional HCs
- Carbon capture & storage

Strategic Aims of Petroleum MSc courses

Develop multi-skilled geoscientists and engineers for the modern petroleum industry

Integrate the core disciplines of geology & petroleum engineering to provide:

- Specialist skills in core subjects
- Competence in allied subjects

Reflect present-day industry structure, workflows and methodologies

Provide students with a framework for pursuing future professional careers

Provide the international petroleum industry with technically competent, welltrained and motivated graduate recruits

Petroleum Geoscience MSc: 52 Students, 15 Countries

Argentina, China, Colombia, Cyprus, France, Greece, India, Indonesia, Kazakhstan, Malaysia, Nigeria, Russia, Saudi Arabia, Spain, UK



Imperial College multi-scale imaging lab

Start with the fundamentals – understand processes experimentally at the pore scale. Micron-to-metre imaging with *in situ* displacement at reservoir conditions.



Dynamic Tomography at Synchrotron Sources



The need for CO₂ storage

- Climate change mitigation: CO₂ storage underground
- Groundwater use and protection
- Geothermal energy
- Oil & gas production
- Shale oil and gas

Carbon capture and storage alone will create an industry the size of the current oil & gas industry, if not larger!



Rogelj et al, Nature Climate Change, 2018

Net Zero by 2050

Our ambition is to be a net zero company by 2050 or sooner. And to help the world get to net zero. This will mean tackling around 415 million tonnes of emissions – 55 million from our operations and 360 million tonnes from the carbon content of our upstream oil and gas production. Importantly these are absolute reductions, to net zero, which is what the world needs most of all. We are also aiming to cut the carbon intensity of the products we sell by 50% by 2050 or sooner

"The world's carbon budget is finite and running out fast; we need a rapid transition to net zero"

Bernard Looney, chief executive officer 13th February 2020



Imperial College London

https://www.theguardian.com/education/2018/nov/22/its-like-tobacco-funding-health-research-should-universities-take-money-from-fossil-fuel

'It's like tobacco funding health research': should universities take money from fossil fuel?

Fossil fuel companies have infiltrated universities for years.

Are students and academics increasingly turning against them?







Trapping, flow and dynamics

There are two things we need to design:

- **1 Trapping.** One phase is surrounded by the other in the wider pore spaces and cannot flow. CO_2 storage, surgical masks.
- **2 Flow.** Both phases can flow over a wide saturation range with little trapping or retention of either phase. Fuel cells, oil recovery.

Controlled by pore structure and *wettability* – the local distribution of contact angle.

Finally, even at low flow rates see the emergence of *non-linear dynamics*.

Trapped CO₂ clusters – colour indicates size

How much is trapped and how much can be stored?

Results in sandstones (Doddington, Bentheimer and Berea).







Consider the fluid-fluid interfaces.

In three dimensions have two principal radii of curvature, r_1 and r_2 , in orthogonal directions: $\kappa_1 = 1/r_1$ and $\kappa_2 = 1/r_2$. Can have different sign. (Total) curvature $\kappa = (\kappa_1 + \kappa_2)$:

Young-Laplace – capillary pressure, $P_c = \sigma \kappa$ Gaussian curvature is defined as $G = \kappa_1 \times \kappa_2$.

Gauss-Bonnet theorem

$$\int GdS = 4\pi\chi$$
 ,

where χ is the Euler characteristic (objects – loops + holes).

A large negative value implies an object with good connectivity (many loops in the structure) while a positive values – ball shaped interfaces – implies poor connectivity.



Trapping and flow, apples and pears

So for trapping, want one phase to be wetting (contact angles below 90°) – the non-wetting phase is trapped. Ball-shaped fluid-fluid interfaces: two approximately equal radii of curvature r_1 and r_2 . $G = 1/r_1r_2 > 0$. Trapped phases.

So what's the opposite? Two approximately *equal and opposite* curvatures: $r_1 = -r_2$. Surely this is ideal for flow/recovery?

Apples and pears!



Minimal surfaces



Minimal surfaces (a catenoid, left, and a helicoid, right) have a negative Gaussian curvature – the product of the two curvatures but a zero mean curvature. G < 0, $\kappa = 0$.

This implies that the fluid phases are well connected.

Minimal surfaces are found in equilibrium when the three-phase contact lines are pinned.

How can we engineer this in porous materials?

Catenoid: $\frac{1}{a^2} \cosh^2 az = x^2 + y^2$



The mixed-wet state

Have a wide range of contact angle – it varies from place to place. Contacts are pinned, as shown.

Rocks in contact with crude oil, soils with organic material, mixture of hydrophilic and hydrophobic fibres.



Minimal surfaces – small curvature



And then....?

Quantify flow properties in rocks: is this really good for recovery?

Measure wettability in different ways – displacement and topology.

Three fluid phases.

Dynamic effects.

And – finally – back to masks and fuel cells.





Favourable oil recovery



Brine saturation increases by water layer swelling.

So, brine bulges into the oil. As the saturation of brine increase these patches of brine expand but are still disconnected

This allows oil to drain to low oil saturation by flow through connected oil layers.

Topology and contact angle

Apply the Gauss-Bonnet theorem. There is a deficit curvature representing the kink at the contact line – can relate this to contact angle.





Phase 2

Phase 2

$$\Delta E = (\Delta A_{12} - \Delta A_{1s} \cos \theta_t)\sigma.$$

Work done – change in Helmholtz free energy (Morrow, 1970):

 θ_{g}

Phase 1

Displacement

 $\theta_g=66^\circ$

Phase 1

$$\Delta F = 0 = \Delta E + P_c \Delta V_1 = \Delta E + P_c V \phi \Delta S_1, \qquad P_c = \sigma \kappa = \sigma \left(\frac{1}{r_1} + \frac{1}{r_2}\right),$$

Final equation – in terms of areas per unit volume, a=A/V:

$$\Delta a_{1s} \cos \theta_t = \kappa \phi \Delta S_1 + \Delta a_{12}$$

Can measure all these terms on pore-space images.

Three-phase flow: Young equations

Bartell-Osterhof relation

$$\sigma_{os} = \sigma_{ws} + \sigma_{ow} \cos \theta_{ow},$$

$$\sigma_{gs} = \sigma_{ws} + \sigma_{gw} \cos \theta_{gw},$$

$$\sigma_{gs} = \sigma_{os} + \sigma_{go} \cos \theta_{go}.$$

$$\sigma_{ws} + \sigma_{gw} \cos \theta_{gw} = \sigma_{os} + \sigma_{go} \cos \theta_{go},$$

 $2_{\sigma_{gw}\cos\theta_{gw}} = \sigma_{ow}\cos\theta_{ow} + \sigma_{go}\cos\theta_{go}.$



Why ducks (and surgeons in masks) don't get wet

Water-wet and oil-wet media: spreading, so $\theta_{qo} = 0$

 $\sigma_{gw}\cos\theta_{gw} = \sigma_{ow}\cos\theta_{ow} + \sigma_{go}\cos\theta_{go}.$ (8.8)

Water-wet: $\cos \theta_{ow} = 1$

 $\cos\theta_{gw} = (\sigma_{ow} + \sigma_{go})/\sigma_{gw} = 1$, as $C_s = \sigma_{gw} - \sigma_{ow} - \sigma_{go} = 0$

Oil-wet: $\cos \theta_{ow} = -1$ $\cos \theta_{gw} = (-\sigma_{ow} + \sigma_{go})/\sigma_{gw} < 0$, as $\sigma_{ow} > \sigma_{go}$

An oil-wet (or made of oil) surface is hydrophobic: repels water – gas is wetting to water.



Energy balance: three-phase flow

Energy balance on pore-scale images is used to find contact angles, θ , in three-phase flow:

 $(\Delta a_{1s} \cos \theta_{12} - \Delta a_{12} - \phi \kappa_{12} \Delta S_1) \sigma_{12} = (\Delta a_{3s} \cos \theta_{23} + \Delta a_{23} - \phi \kappa_{23} \Delta S_3) \sigma_{23} + \Delta a_{13} \sigma_{13}$

 Δ is the difference in specific interfacial areas *a*, and saturations *S*. σ is interfacial tension, κ total curvature, and ϕ porosity. 1, 2, 3 and *s* label the three fluid phases and the solid respectively.

Phase 3, Gas

Phase 2, Oil

Phase 1, Water

Rock



$$\theta_{12} = 48^{\circ}, \theta_{13} = 44^{\circ}, \theta_{23} = 0^{\circ}$$

Water is wetting, oil is intermediate-wet and gas is non-wetting

 $\theta_{12} = 134^o$, $\theta_{13} = 119^o$, $\theta_{23} = 66^o$

Altered-wettability

Oil is wetting, gas is intermediate-wet and water is non-wetting

Blunt et. al. JCIS (2021)

Dynamic effects – non-Darcy flow

Measure pressure drop as a function of flow rate (capillary number): see the emergence of non-linear behaviour. Study different fractional flows in steady state.



Can predict the onset of intermittency



The phase diagram of the transition from Darcy flow (empty symbols) to intermittent flow (filled symbols) as a function of non-wetting phase capillary number Ca_{nw} and wetting phase capillary number Ca_{w}

Threshold line equation using simple energy balance:

$$Ca_{nw}^{i} = Y^{i}(1 - f_{w})^{2}$$
$$Ca_{w}^{i} = Y^{i}f_{w}(1 - f_{w})\frac{\mu_{w}}{\mu_{nw}}$$

$$Y^i = \frac{\mu_{nw}}{\mu_w} \frac{Kr^2}{\phi l^4}$$

Details can be found:

Zhang, Y., Bijeljic, B., Gao, Y., Lin, Q., & Blunt, M. J. (2020, August 20). Quantification of non-linear multiphase flow in porous media. https://doi.org/10.31223/osf.io/2rxbn

Works on literature data



- Gao et al. (2017) (Bentheimer, decane/brine) Intermittent flow
- ◄ Gao et al. (2019) (Estaillades, decane/brine) Intermittent flow
- Spurin et al. (2019b) (Estaillades, decane/brine) No intermittent flow
- Spurin et al. (2019b) (Estaillades, nitrogen/brine) Intermittent flow
- ★ Spurin et al. (2019b) (Estaillades, hexadecane/brine) No intermittent flow
- * Datta et al. (2014) (Glass beads, five cases) Fixed pathways
- * Datta et al. (2014) (Glass beads, five cases) Intermittent and disconnected ganglia

We apply the theory successfully to literature data.



Spurin et. al. PRE (2019)

Surgical masks and fuel cells

Masks: porous fibrous plastic (oil-wet) layer to repel/trap water.

Aerosols are smaller than the pore size, so need to stick to the surface. Have a water-wet (retains water) layer.

So alternating layers of hydrophobic and hydrophilic layers.

Can we design this more effectively?

Fuel cells: mix of naturally water-wet carbon fibres and Teflon-coated coated fibres (hydrophobic) – mixed-wet.

Again can we design this to maximize flow?





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TOTAL











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Rock Technologies