

#### Joel Voldman

#### **Massachusetts Institute of Technology**

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#### Outline

- > Review of last time
- > Poiseuille flow
- > Stokes drag on a sphere
- > Squeezed-film damping
- > Electrolytes & Electrokinetic separations

## Last time



#### Outline

- > Review of last time
- > Poiseuille flow
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- > Squeezed-film damping
- > Electrolytes & Electrokinetic separations

- > Pressure-driven flow through a pipe
  - In our case, two parallel plates
- > Velocity profile is parabolic
- > This is the most common flow in microfluidics
  - Assumes that h<<W</p>



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Adapted from Figure 13.5 in Senturia, Stephen D. Microsystem Design. Boston, MA: Kluwer Academic Publishers, 2001, p. 329. ISBN: 9780792372462.

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# **Solution for Poiseuille Flow**

#### > Assume

- Incompressible
- Steady
- U<sub>x</sub> only depends on y
- Ignore gravity
- > Assume a uniform pressure gradient along the pipe
- > Result is Poisson's eqn
- > Boundary conditions:
  - Relative velocity goes to zero at the walls
    - » no-slip boundary condition

$$\rho_{\rm m} \left( \frac{\partial \mathbf{U}}{\partial t} + \mathbf{U} \cdot \nabla \mathbf{U} \right) = -\nabla P + \eta \nabla^2 \mathbf{U} + \frac{\eta}{3} \nabla \left( \nabla / \mathbf{U} \right) + \rho_{\rm m} \mathbf{g}$$

$$\frac{dP}{dx} = -K$$
$$\frac{\partial^2 U_x}{\partial y^2} = -\frac{K}{\eta}$$

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# **Solution for Poiseuille Flow**

- Integrate twice to get solution
- > Maximum velocity is at center
- > Can get linear flowrate [m/s] and volumetric flowrate [m<sup>3</sup>/s]
- > Can get lumped resistor using the fluidic convention
- > Note STRONG dependence on h
- > This relation is more complicated when the aspect ratio is not very high...



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$$U_{x} = \frac{1}{2\eta} \left[ y \left( h - y \right) \right] K$$

$$U_{\rm max} = \frac{h^2}{8\eta} K$$

$$Q = W \int_{0}^{h} U_{x} dy = \frac{Wh^{3}}{12\eta} K$$

$$\Delta P = \text{effort} = KL$$

$$\Delta P = \frac{12\eta L}{Wh^3}Q$$

$$\Rightarrow R_{Pois} = \frac{12\eta L}{Wh^3}$$

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- It takes a certain characteristic length, called the development length, to establish the Poiseuille velocity profile
- > This development length corresponds to a development time for viscous stresses to diffuse from wall
- > Development length is proportional to the characteristic length scale and to the Reynolds number, both of which tend to be small in microfluidic devices

time 
$$\approx \frac{L^2}{\eta^*} \approx \operatorname{Re} \frac{L}{U}$$
  
 $L_D \approx (time)U \approx \operatorname{Re} L$ 

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# A note on vorticity

- > A common statement is to say that laminar flow has no vorticity
- > What is meant is that laminar flow has no turbulence
- > Vorticity and turbulence are different
- > Can the pinwheel spin?
  - Then there is vorticity
- > Demonstrate for Poiseuille flow

 $\boldsymbol{\omega} = \nabla \times \mathbf{U}$ Vorticity



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#### **Stokes Flow**

- Steady-state flow in which inertial effects can be neglected, Re→0
- > The result is a vector Poisson equation
- > Also called "creeping flow"
- > Action is instantaneous
  - No "mass" in system
  - Incompressible: no springs
- > This is a typical approximation made in microfluidics

When  $\rho_m \frac{DU}{Dt}$  can be neglected

$$\eta \nabla^2 \mathbf{U} = \nabla P^*$$

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#### Stokes' drag on a sphere

- In creeping flow, one can solve for the flow field around a sphere placed in an initially uniform flow field
- > This can be used to find the stresses on the sphere and sum them to find the total drag
- > This is called the Stokes' drag
- > This is often the predominant particle force in microfluidic systems
- > See Deen's text for derivation



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$$F_d = 6\pi\eta R U$$

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# Stokes' drag on a sphere

- > This is strictly valid ONLY in a uniform flow
- > These are hard to make...
- Instead, we can use U<sub>x</sub>(y) of the parabolic flow profile to calculate a height-dependent drag force
- > This approach fails when the particle is too big
- Instead, take advantage of published solutions
  - Shear flow: Goldman et al., Chem. Eng. Sci. 22, 653 (1967).
  - Poiseuille flow: Ganatos et al. *J. Fluid Mech.* 99, 755 (1980).



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#### **Squeezed-Film Damping**

- > This is how we will get our *b* (or *R*) for the parallel-plate actuator
- > The result of motion against a fluid boundary
  - If the fluid is incompressible, there can be a large pressure rise, so large back forces result
  - If the fluid is compressible, it takes finite motion to create a pressure rise
- In either case, the dissipation due to viscous flow provides a damping mechanism for the motion
- > This is related to "lubrication theory"



MA: Kluwer Academic Publishers, 2001, p. 333. ISBN: 9780792372462.

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# **The Reynolds Equation**

- Assumptions for compressible isothermal squeezed-film damping
  - One-dimensional pressure gradient: P(r,t) = P(y,t) only
    - » No pressure gradient in z or along plate (x)
  - Stokes flow
  - Poiseuille flow profile in the plane
  - Ideal gas law
  - Isothermal (temperature rise due to compression is small, and heat flow to the walls is rapid)
  - No-slip BC's
  - Rigid plate: h(r,t) = h(t) only

$$\frac{\partial(Ph)}{\partial t} = \frac{h^3}{12\eta} \left(\frac{1}{2}\nabla^2 P^2\right)$$



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Adapted from Figure 13.7 in Senturia, Stephen D. *Microsystem Design*. Boston, MA: Kluwer Academic Publishers, 2001, p. 334. ISBN: 9780792372462.

#### **Example: Rigid Plate Damping**

> Now, assume small motions

→ Linearize

> The result is (guess what!) the heat-flow equation

$$\frac{\partial(Ph)}{\partial t} = \frac{h^3}{12\eta} \left(\frac{1}{2}\nabla^2 P^2\right)$$



Image by MIT OpenCourseWare. Adapted from Figure 13.7 in Senturia, Stephen D. *Microsystem Design*. Boston, MA: Kluwer Academic Publishers, 2001, p. 334. ISBN: 9780792372462. If we linearize:  $h = h_0 + \delta h$   $P = P_o + \delta P$ Normalize:  $\xi = \frac{y}{W}$   $\hat{p} = \frac{\delta P}{P_o}$ 

$$\frac{\partial \hat{p}}{\partial t} = \frac{h_0^2 P_o}{12\eta W^2} \frac{\partial^2 \hat{p}}{\partial \xi^2} - \frac{\dot{h}}{h_o}$$

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#### **Suddenly Applied Motion**

- > We already solved a very similar problem
  - Impulse of heat into 1-D resistor
- Now we have a velocity impulse: sudden change of height
- > We get a series of 1<sup>st</sup>-order terms, as before
- > Only need 1<sup>st</sup> term, which is an RC circuit
- > R for viscous damping
- > C for gas compressibility
- > Details are in the book

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#### **Electrokinetic Phenomena**

- It is a coupled-domain problem in which electrostatic forces result in fluid flow (and vice versa)
- Start with electrolytes, move into double layer, and finally show how to manipulate the double layer
- > It is the driver behind ALOT of early micro-TAS work

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### **Electrolytes**

- > Electrolytes: liquids with mobile ions
  - Examples: water, PBS
- > lons can move via concentration gradients (diffusion) or electric fields (drift)
- Macroscopically, the liquid is approximately charge-neutral (called quasineutrality)



$$D_e = \sum_i z_i q_e C_i \approx 0$$
 in the bulk

In neutral regions:  $\nabla^2 \phi = \frac{-\rho_e}{\varepsilon} = 0$ 

For a binary electrolyte \_ (e.g., NaCl in water)

$$N_{+} = u_{+}C_{+}E - D_{+}\nabla C_{+}$$
  

$$N_{-} = -u_{-}C_{-}E - D_{-}\nabla C_{-}$$
  

$$\rho_{e} = q_{e}(C_{+} - C_{-}) \approx 0$$

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# **Electrolytes**

- Surfaces with fixed charge can lead to net space charge in the liquid
- Diffusion competes with drift, and at equilibrium, Boltzmann distribution follows
- > This leads to the Poisson-Boltzmann equation



$$N_{i} = \frac{z_{i}}{|z_{i}|} u_{i}C_{i}E - D_{i}\nabla C_{i} = 0 \text{ at equilibrium}$$
$$\Rightarrow C_{i}(x) = C_{i,o}e^{-\frac{z_{i}q_{e}(\phi(x) - \phi_{o})}{k_{B}T}}$$

Near the wall: 
$$\nabla^2 \phi = -\frac{\rho_e}{\varepsilon} \implies \nabla^2 \phi = -\frac{1}{\varepsilon} \sum_i z_i q_e C_{i,o} e^{-\frac{z_i q_e}{k_B T}}$$

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#### **Diffuse Double Layer**

 $\hat{\phi}$ 

 $\phi_w$ 

 $L_D$ 

Express in terms of 
$$\hat{\phi} = \phi - \phi_0$$
:  $\nabla^2 \hat{\phi} = -\frac{1}{\varepsilon} \sum_i z_i q_e C_{i,o} e^{-\frac{z_i q_e \phi}{k_B T}}$ 

Expand for small potential variations

$$\nabla^2 \ \hat{\phi} \cong -\frac{1}{\varepsilon} \sum_i z_i q_e C_{i,o} + \frac{q_e^2}{\varepsilon k_B T} \left[ \sum_i z_i^2 C_{i,o} \right] \hat{\phi}$$

Assuming the reference region is charge neutral



For binary monovalent electrolyte (e.g., NaCl)



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# > We can calculate the total charge in the double layer > It must balance the charge at the wall



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#### **Actual Double Layers**

- > The actual situation is MUCH more complicated
- > Some ions are tightly or specifically adsorbed, forming the Stern layer and screening the wall charge
- > The rest distribute in a diffuse double-layer: the Gouy-Chapman layer
- > This is an active area of research



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#### **Electroosmotic Flow**

#### > An axial electric field exerts a force on the charge in the diffuse double layer, which drags the fluid down the pipe



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#### **Analysis of Electroosmotic Flow**

#### > Assume

- Creeping flow
- One dimensional flow  $U_x(z)$
- No pressure drop
- Electrical body force
- > Express charge density in terms of wall surface charge density
- > Set up differential equation



#### **Analysis of Electroosmotic Flow**



 $\sim 3L_D$ 



Integrate twice, and use boundary conditions:



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## **Analysis of Electroosmotic Flow**

- > This leads to a PLUG-FLOW profile
- > Flow depends on zeta

potential  $\zeta$ 

- The potential at the slip plane  $\delta$
- Which is in a different place than the wall, the Stern layer, or  $L_{\rm D}$
- It is what is measured experimentally
- For  $h >> L_D$ , one typically assumes

5

$$\delta = 0$$
$$\phi_w = \zeta$$

$$U_{x} = -\frac{\varepsilon}{\eta} \left( \zeta - \hat{\phi} \right) E_{x}$$

$$U_{0} = -\frac{\sigma_{w}L_{D}}{\eta}e^{-\frac{\delta}{L_{D}}}E_{x} = -\frac{\varepsilon\zeta}{\eta}E_{x}$$
  
for  $z > 3L_{D}$ 

## Electrophoresis

- > This is just electroosmosis around a solid surface
- > Each ionic species has its own mobility
- > Therefore, in an electrolyte in which there is a net electric field, ions will drift at various rates
- > This is the basis of a separation technology called electrophoretic separation



## **Electrokinetic separation**

- > This was THE original driver for micro-TAS
  - TAS = total analysis systems
- > Create fully integrated microsystems that would go from "sample to answer"
- > The KEY enabler was the integration of nonmechanical valves with the separation column
  - This creates extremely narrow sample plugs
- > Also important is the ability to multiplex

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- > The actual sample prep was (and is) usually ignored
- > This is/was the raison d'être of Caliper & Aclara
- > Aclara died, unclear if Caliper will succeed

## **Electrokinetic separation**

- > We use a channel-crossing structure to select a sample plug
- > Then we switch voltages to drag the plug down a separation column
- In one approach, EO and EP coexist, but EO dominates
- Different species travel at different rates separation



$$L_{s} = t_{sep} \Big| \mu_{EP,1} - \mu_{EP,2} \Big| \mathsf{E}_{x}$$

#### **Separation time**

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#### **Schematic Illustration**

- > The key discovery was that liquid samples could be controlled with voltages
- > This allows one to valve and pump liquids
  - Create small sample plugs



#### **ORNL** movie

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#### **Electrokinetic separation**

- > The "macro" technology is "conventional" capillary electrophoresis
  - Most notably used in the human genome project
- Sample loading is the problem
- > To separate two species (ignoring diffusion), we need
- Smaller starting W means we can use a shorter channel
- > And get a faster separation



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# **Electrokinetic separation**

- > Conventional capillaries have larger W because injection is not integrated
  - Though this is always getting better
- > This thus requires a longer channel
- > Microfab also allows for integration



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#### An aside on electrodes

- > Current in the electrolyte is carried by ions
- > Current in the wire is carried by electrons
- > At the surface, something must happen to transfer this current
- > This is electrochemistry and the typical byproduct are gases → bubbles

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#### **Electrokinetics**

#### > That is just the beginning

#### > Dielectrophoresis

- Force on dipoles in non-uniform electric fields
- Can use AC fields
- Can hold things in place

#### > Other phenomena

- Electrohydrodynamics
- Electrowetting
- Induced-charge electrophoresis/electroosmosis

If you are designing a chip that needs to move liquids around, which method is best?

#### > Issues to consider

- Flowrate scaling
- Liquid composition
- System partitioning
- Materials
- Species Transport

#### > Flowrate

- Water in rectangular SiO<sub>2</sub> channel
- h varies, W=1000 μm, L=2 cm, ε=80ε<sub>0</sub>, ζ=50 mV
- Drive with  $E_x$ =100 V/cm,  $\Delta$ P=5 psi



#### > Scaling

- Pressure-driven flow larger in large channels
  - » Due to cubic dependence of flow resistance
- EO flow larger in small channels
- > Both scale equivalently with channel length
  - Larger *L* requires more voltage and higher pressure to get same flow



- > Other issues typically matter more
- > Valving
  - EOF has "built-in" valving using E-fields
  - Poiseuille needs mechanical valves
- > Liquid limitations
  - Poiseuille flow can pump any liquid
  - EOF ionic strength limits
    - » Debye length depends on  $1/\sqrt{C_0}$
    - » Increasing ionic strength decreases  $L_D$  and thus EOF
    - » Typically use ~10-100 mM salt buffer
  - EOF pH limits
    - » pH affects wall charge → affects EOF
    - » Typically use pH ~7 buffers

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#### > Materials issues

- Poiseuille flow can use any material
- EOF requires defined (and stable) surface charge
  - » Best is silica (or at least glass)
  - » Polymers are more difficult
  - » Surface charge can change as molecules adsorb, etc.
- > System partitioning
  - Both approaches involve off-chip components
    - » Electrodes for EOF
    - » Pumps for Poiseuille flow
  - Integrating complete lab-on-a-chip usually BAD idea
    - » EOF: Easier to use external electronics
    - » Poiseuille: Pumps hard to make on-chip

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#### > Transport limitations

- EOF
  - » Will separate molecules as they are convected downstream
  - » Species in flow must tolerate E-fields (DNA/proteins OK, cells not so good)
  - » Plugs remain plugs
- Poiseuille flow
  - » Will not separate molecules
  - » Objects in flow must tolerate shear/pressure (DNA/proteins OK, cells OK depending on shear/pressure)
  - » Will distort plugs

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#### What's next

- > We will discuss the behavior of the stuff in the liquids
- > How to manipulate that stuff at the microscale
- > And then we head into system-level issues
  - Feedback, Noise, etc.

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