

# Flow patterns induced by hydrophilic surfaces

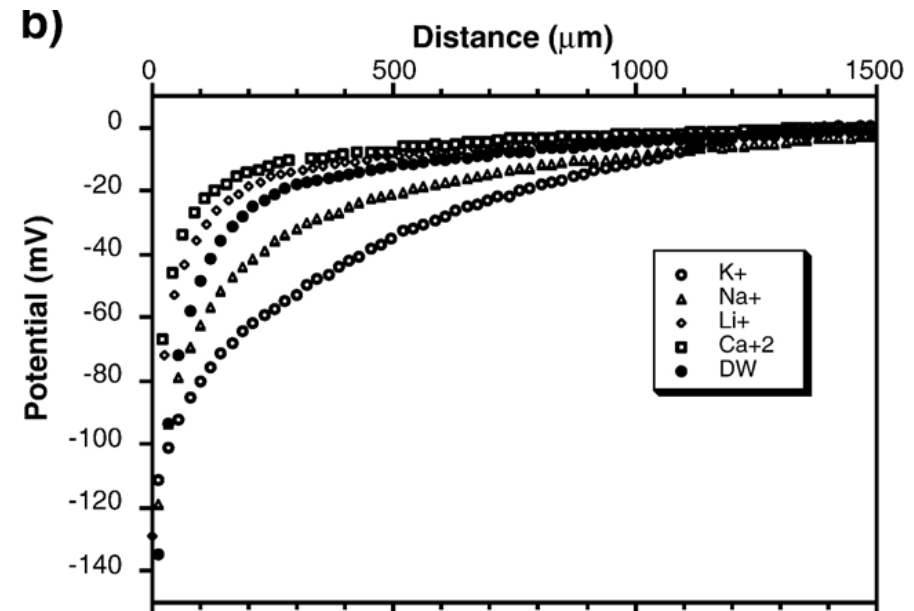
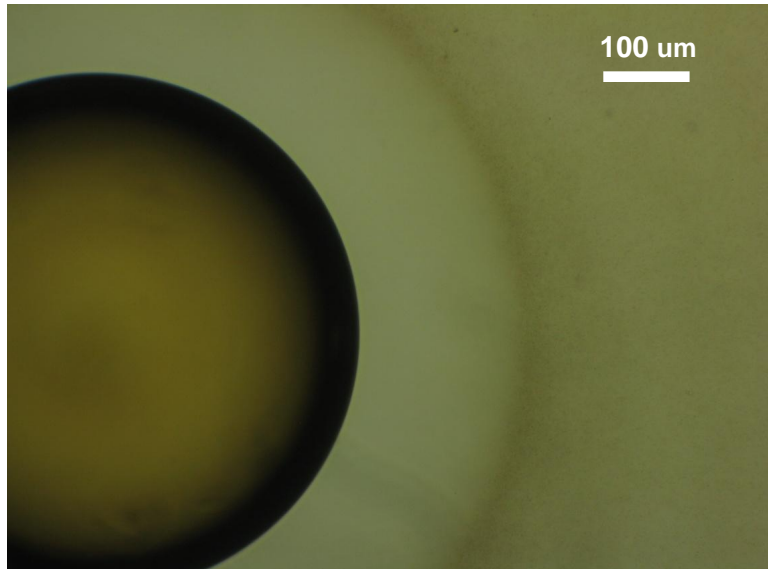
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**Where innovation starts**

# Exclusion zone



- Charged resin bead excluding suspension of microspheres to a distance of > 200 μm. The resulted exclusion zone is surprisingly much larger than the prediction of the double layer theory being ~1 μm.
- A potential gradient is also observed however across the exclusion zone.

# Old references on exclusion zones

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## Modern State of the Investigation of Long-Range Surface Forces\*

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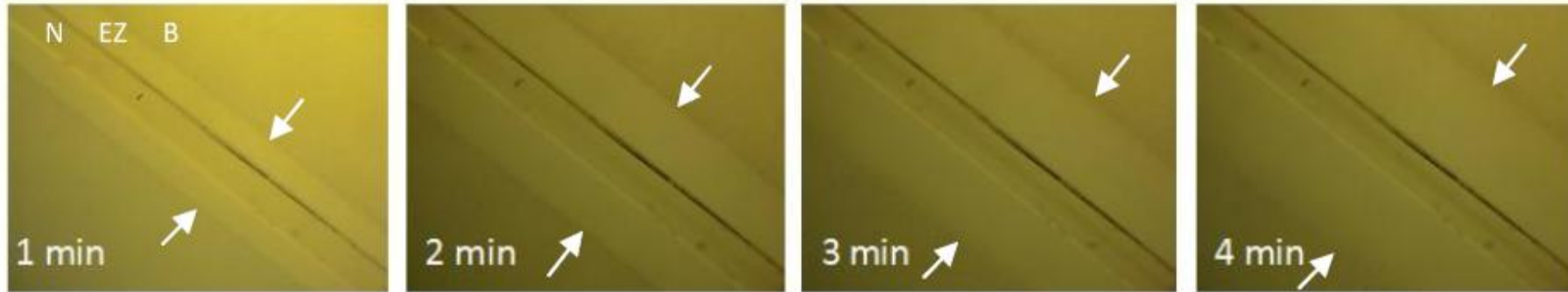
*Received January 15, 1987*

Development of the concept of surface long-range forces and, in particular, the equilibrium disjoining pressure of liquid and gaseous interlayers has been set forth. Considered are the molecular, adsorption, electrical, structural, and electronic components of disjoining pressure. The contribution of the disjoining pressure to the hydrodynamics of thin layers is considered. The first theory of the frost heaving of soils has been formulated. Stated are the investigations of surface forces, in particular, in the processes of the formation of new interfaces and arising phenomena of the emission of electrons, ions, photons, and neutrons.

Up to now, investigation of surface forces has allowed the long-range action to be detected, which does not exceed a micrometers. Therefore, special attention should be paid to the experiments carried out by a researcher of the Oncology Center of the Academy of Medicinal Sciences, Golovanov. Diluting the blood plasma by a 4-fold volume of a 10% salt solution, Golovanov has detected that leukocytes can repel the surrounding erythrocytes at a distance up to 80  $\mu\text{m}$ .<sup>43</sup> It has also been shown that in the absence of erythrocytes, leukocytes that mutually repel one another form in a few hours a periodical lattices possessing hexagonal symmetry.<sup>43,44</sup> In this case, the distance between the neighboring lattice sites occupied by leukocytes was on the order of about 60-70  $\mu\text{m}$ . These investigations results unambiguously indicate that live cells are able to generate repulsion forces having a radius of action which many times exceeds that of the forces stemming from unorganized matter. An attempt was made to ascribe the forces due to leukocytes to diffusion fluxes resulting from live activity processes.

However, this explanation became invalidated when it was demonstrated that the repulsion forces are able to overcome material barriers in the form of filaments and wires.<sup>45</sup> This indicates that the nature of these forces is cognate with that of Frohlich's forces generated by coherent excitations,<sup>46</sup> emitting electromagnetic waves having the length of a few hundred micrometers. The next problem to be solved resides in finding a direct substantiation of this hypothesis.

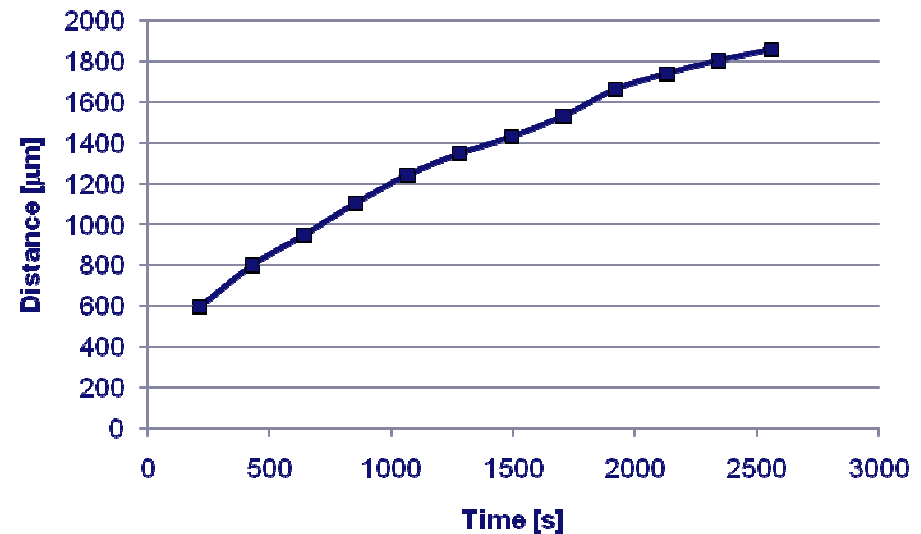
# Time evolution of exclusion zone



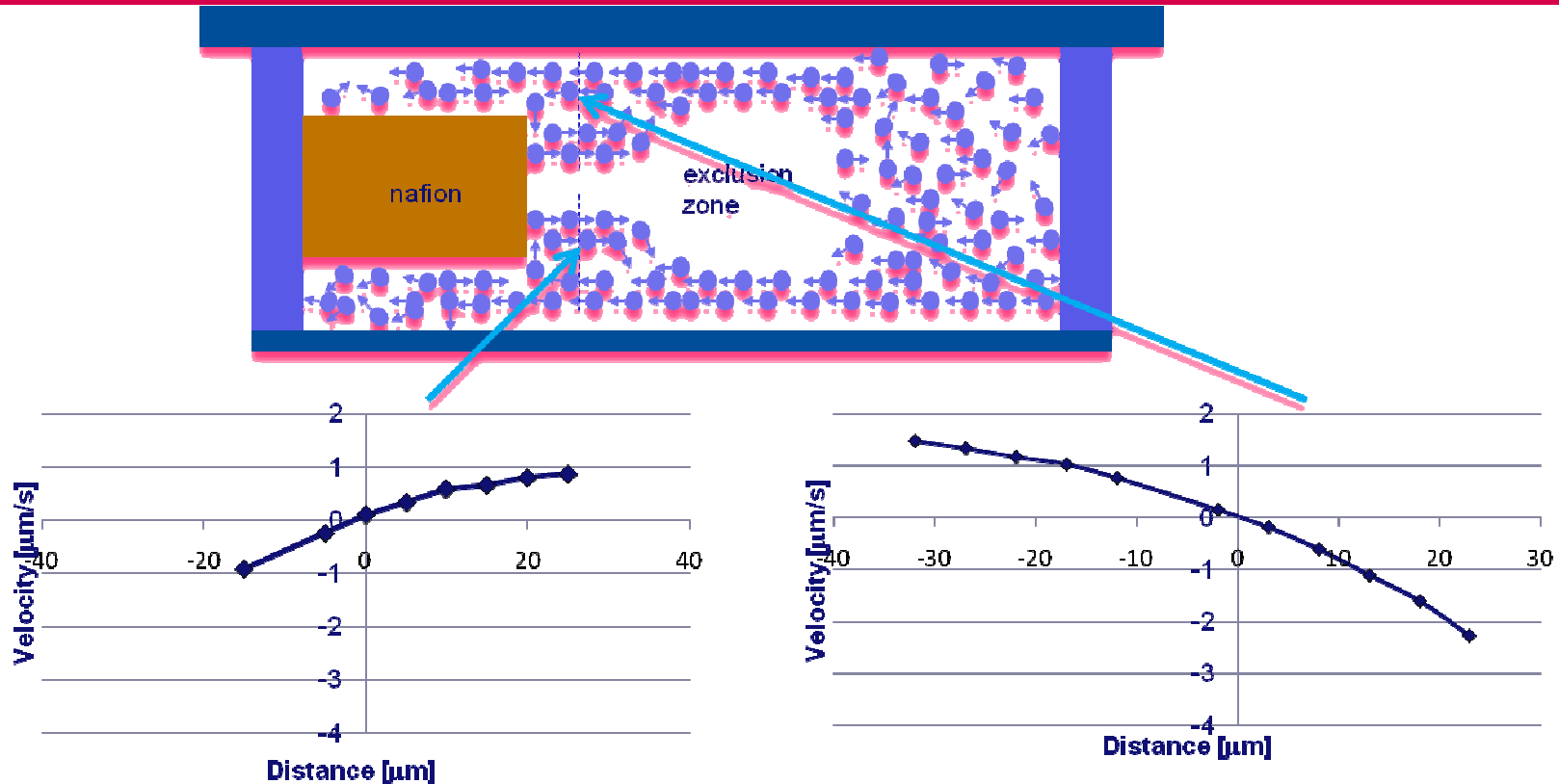
Top: N = Nafion, thickness 200  $\mu\text{m}$   
EZ = exclusion zone  
B = beads ( $\varnothing$  1.0  $\mu\text{m}$ )

1. The exclusion zone expands as a function of time
2. The width of the exclusion zone is proportional to the square root of time

Zone width  $\propto \sqrt{\text{time}}$



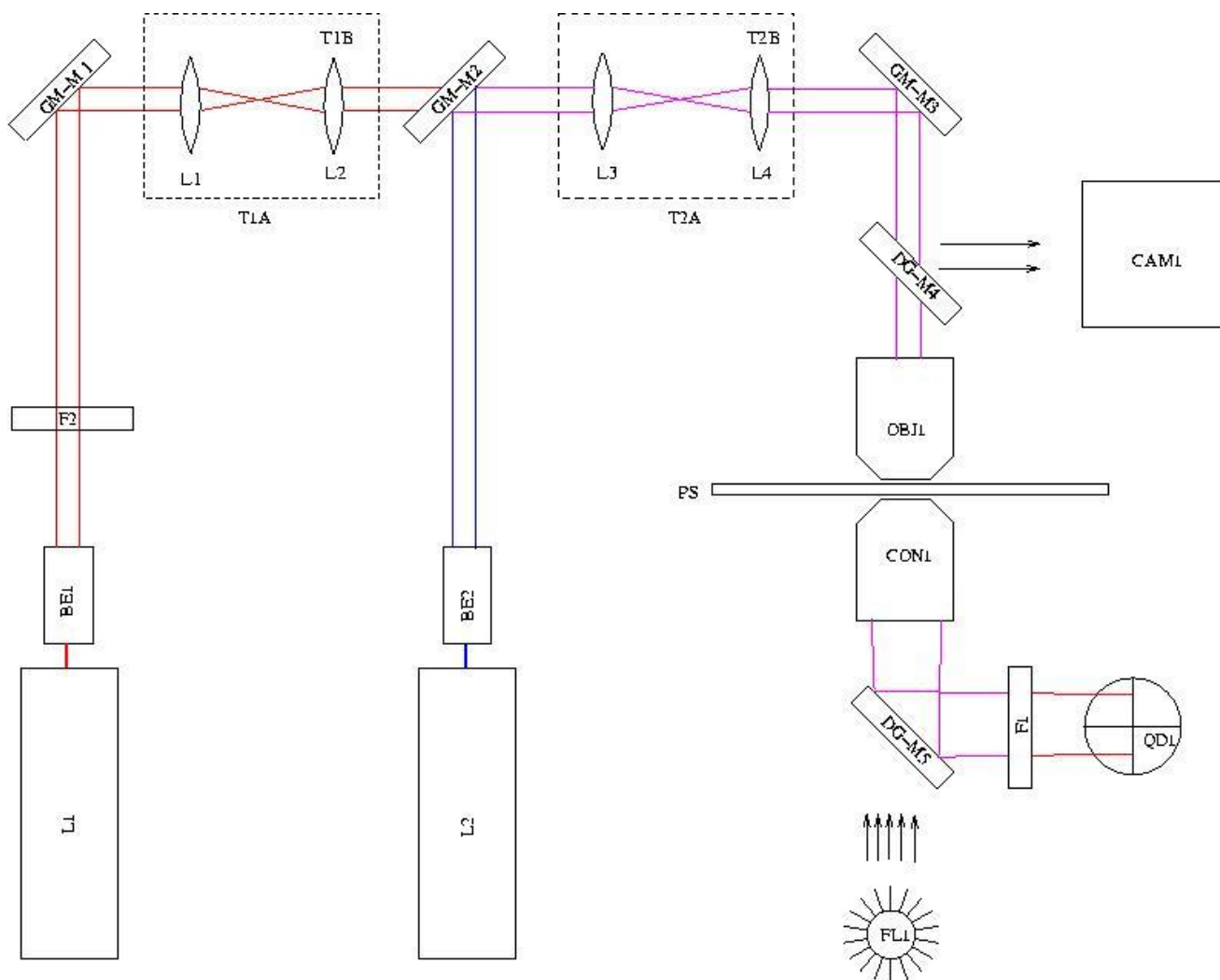
# Counter flow



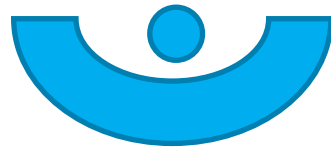
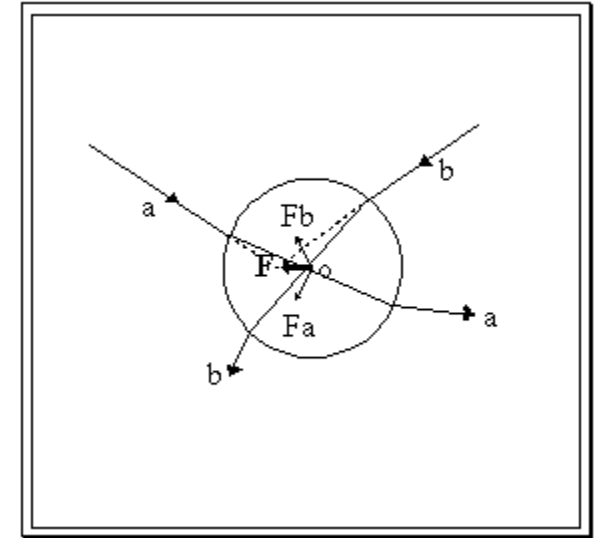
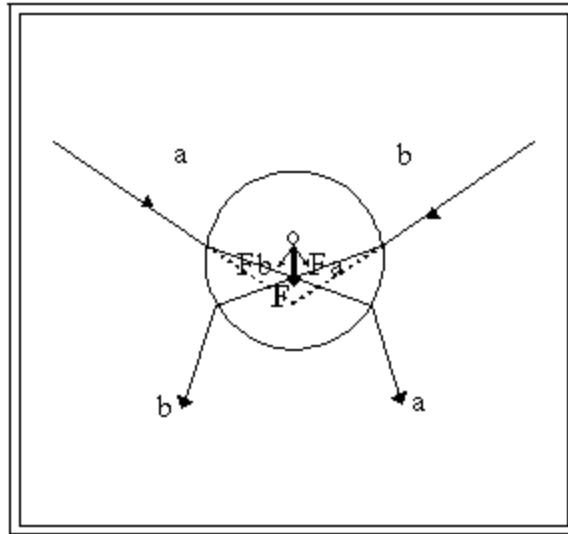
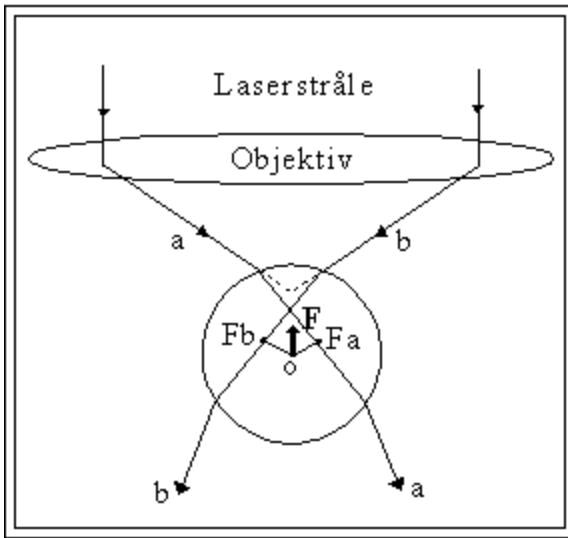
Beads flow from outside the EZ towards the beads creating vortices near the nafion surface

S. Musa et al., submitted to Physical Review E

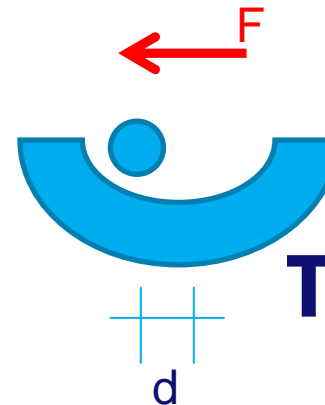
# Viscosity of EZ-water: Optical tweezing



# Viscosity of EZ-water: Optical tweezing



Potential well



# Viscosity

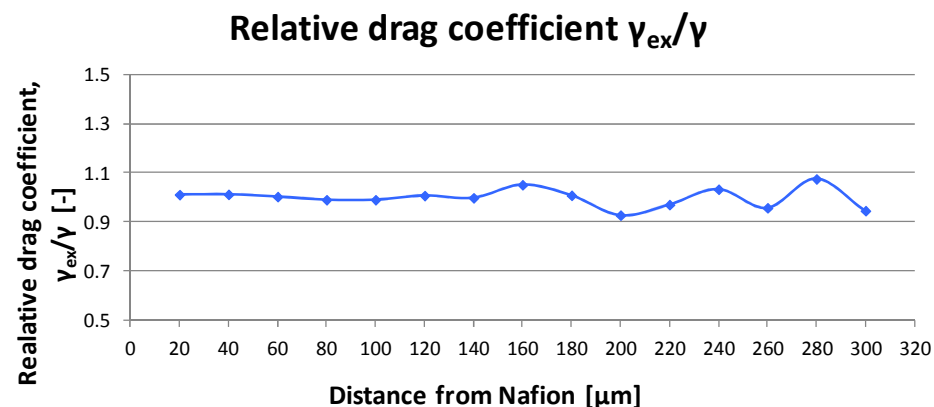
- **3 methods:**
- **Measuring stokes drag under sinusoidal movement of the bead**
- **Measuring Brownian motion of the bead**
- **Measuring Brownian motion of the bead while the bead is moving sinusoidally.**



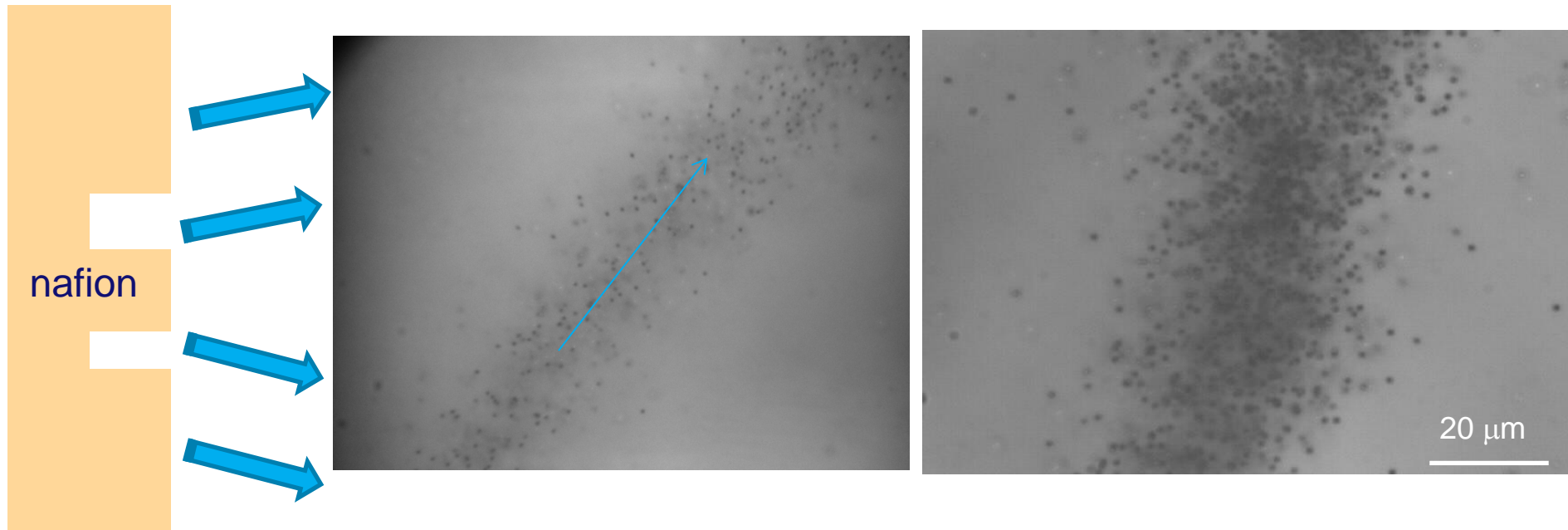
# Viscosity of the EZ-water

We have conducted experiments to examine existence of ordered layer next to the surface by

1. Measuring whether there is a change in viscosity inside and outside the exclusion zone
2. Measuring whether there is a change in optical density inside and outside the exclusion zone

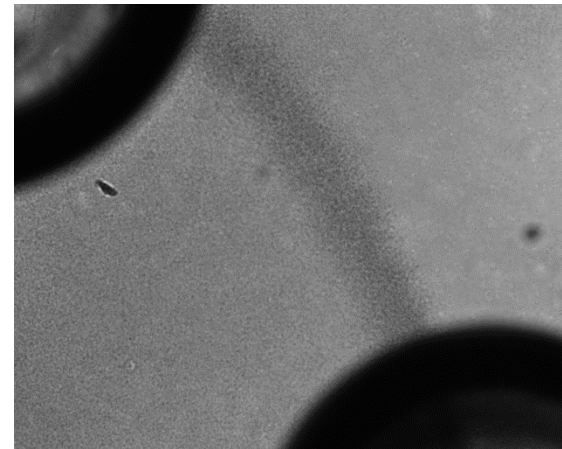
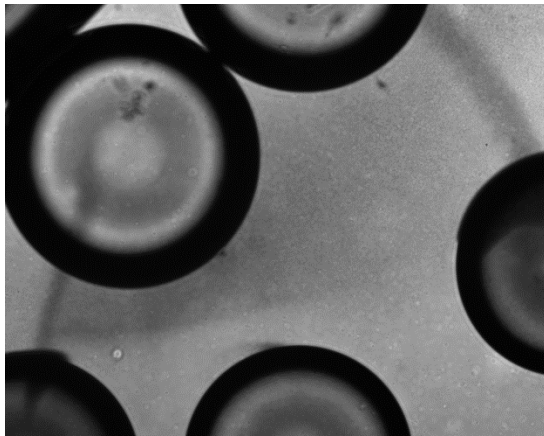


# Applications: confined flow generation



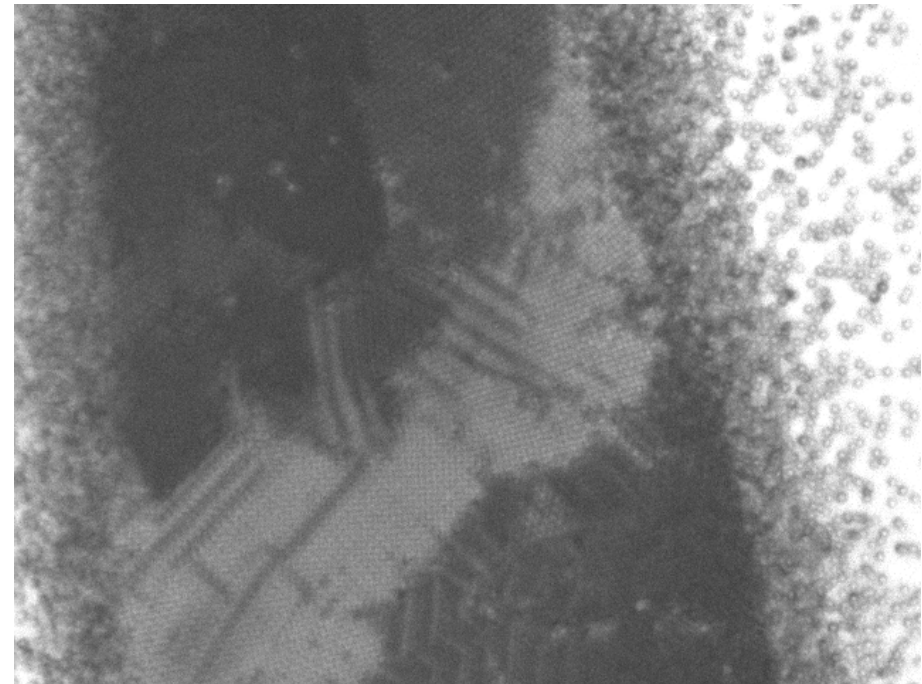
1. When the edge of the Nafion film is structured confined flow lines of bead can be generated with different beads velocities
2. The flow which continues for several hours can be used in application where slow flow generation is needed

# Flow between surfaces of different polarity



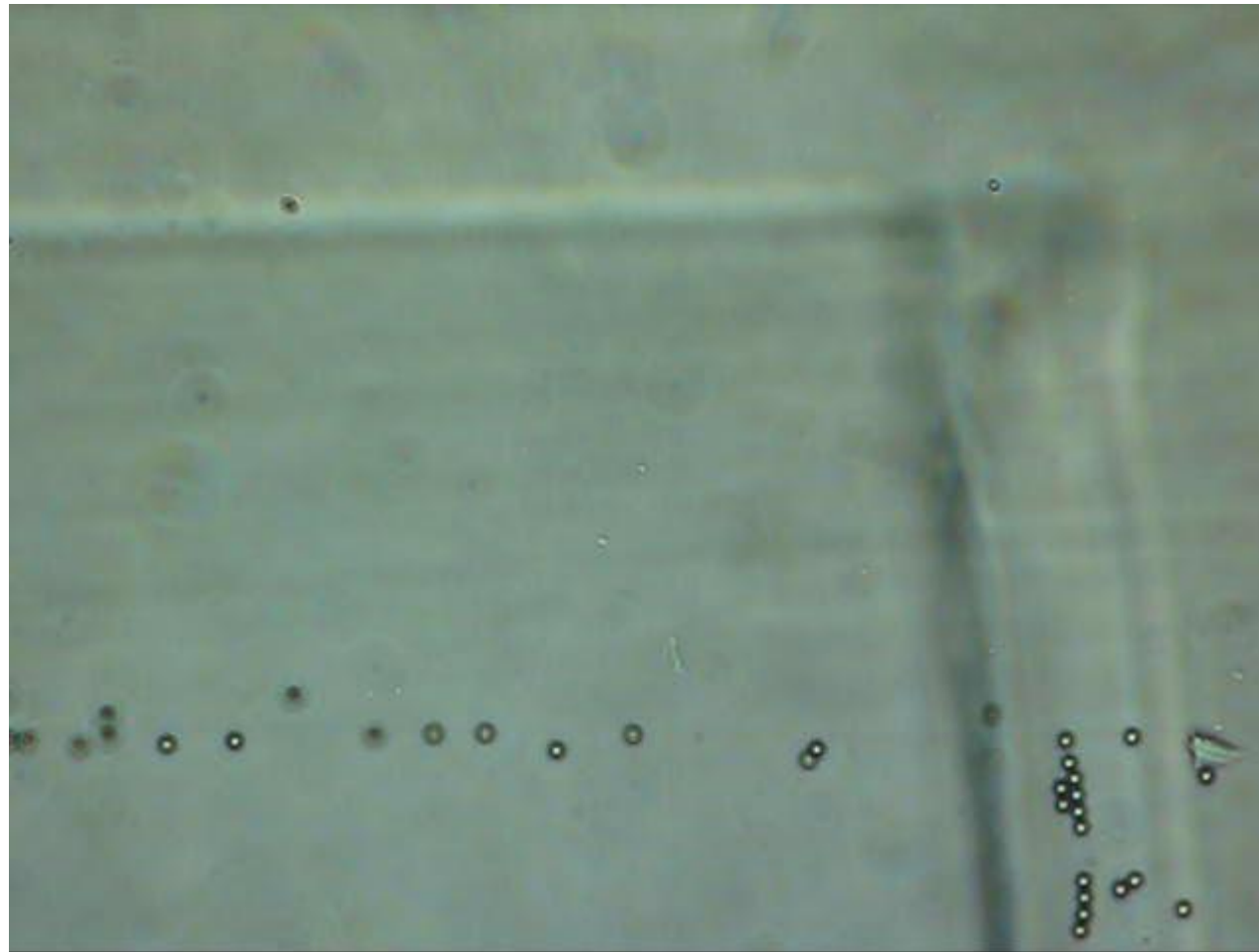
Beads flow between resins with different charged polarities

# Colloidal crystal formation



Large crystals of micro-spheres could form between hydrophilic surface and gloss.

# Colloidal crystal formation



# Possible causes of EZ formation

1. Crystal formation in the EZ-zone
2. Diffusiophoresis moves the beads away from the nafion and causes the vortices.
3. Frohlich coherence mechanism/Coherent domains

# Diffusiophoresis

- **What is diffusiophoresis?**
- **It is the movement of a colloidal particle as a consequence of a concentration gradient of a solute**

# Diffusiophoresis

## Diffusiophoresis and electrophoresis of a charged sphere perpendicular to two plane walls

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### Abstract

The problem of diffusiophoretic and electrophoretic motions of a dielectric spherical particle in an electrolyte solution situated at an arbitrary position between two infinite parallel plane walls is studied theoretically in the quasisteady limit of negligible Peclet and Reynolds numbers. The applied electrolyte concentration gradient or electric field is uniform and perpendicular to the plane walls. The electric double layer at the particle surface is assumed to be thin relative to the particle radius and to the particle-wall gap widths, but the polarization effect of the diffuse ions in the double layer is incorporated. To solve the conservative equations, the general solution is constructed from the fundamental solutions in both cylindrical and spherical coordinates. The boundary conditions are enforced first at the plane walls by the Hankel transforms and then on the particle surface by a collocation technique. Numerical results for the diffusiophoretic and electrophoretic velocities of the particle relative to those of a particle under identical conditions in an unbounded solution are presented for various cases. The collocation results agree well with the approximate analytical solutions obtained by using a method of reflections. The presence of the walls can reduce or enhance the particle velocity, depending on the properties of the particle-solution system and the relative particle-wall separation distances. The boundary effects on diffusiophoresis and electrophoresis of a particle normal to two plane walls are found to be quite significant and complicated, and generally stronger than those parallel to the confining walls.  
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**Keywords:** Diffusiophoresis; Electrophoresis; Boundary effects; Thin but polarized double layer

### 1. Introduction

The electrokinetic transport phenomena of charged colloidal particles suspended in electrolyte solutions are of much fundamental and practical interest in the fields of chemical, biological, and electronic engineering and science. Electrophoresis, which results from the interaction between an applied electric field and the electric double layer surrounding a charged particle, is the most familiar example of various electrokinetic transport phenomena. The electrophoretic velocity  $\mathbf{U}^{(0)}$  of a uniformly charged particle in an unbounded ionic solution is simply related to the uniformly imposed electric field  $\mathbf{E}^\infty$  by the Smoluchowski equation [1–4],

$$\mathbf{U}^{(0)} = \frac{\varepsilon \zeta}{\eta} \mathbf{E}^\infty, \quad (1)$$

In this equation,  $\varepsilon$  and  $\eta$  are the dielectric permittivity and viscosity, respectively, of the fluid, and  $\zeta$  is the zeta potential at the particle surface.

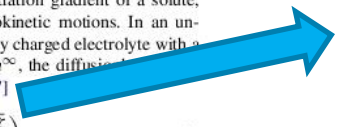
Diffusiophoresis, which is the movement of a particle in response to the macroscopic concentration gradient of a solute, can be another example of electrokinetic motions. In an unbounded solution of a symmetrically charged electrolyte with a constant concentration gradient  $\nabla n^\infty$ , the diffusiophoretic velocity of a dielectric particle is [5–7]

$$\mathbf{U}^{(0)} = \frac{\varepsilon \zeta k T \nabla n^\infty}{Z e n^\infty(\mathbf{0})} \left( \alpha + \frac{\ln \cosh \zeta}{\zeta} \right), \quad (2)$$

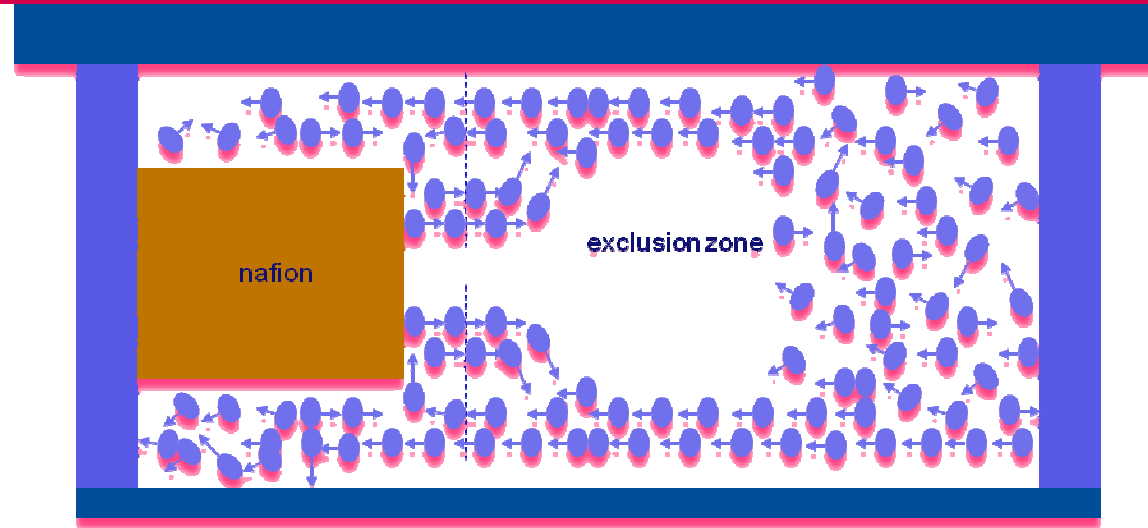
with the dimensionless parameters

$$\alpha = \frac{\tilde{D}_2 - \tilde{D}_1}{\tilde{D}_2 + \tilde{D}_1}, \quad (3)$$

$$\nabla \log c = \frac{\nabla c}{c}$$







Hexagonal water crystal formation. (Pollack)

Hexagonal Bilayer Water tends to dissociate  $\text{OH}^-$  (Anick)

Crystalline scaffold becomes positively charged and therefore sticks to the Nafion.

Crystalline scaffold is saturated with negatively charged liquid water.

The  $\text{OH}^-$  ions move away from the Nafion because of electrophoresis.

The liquid water is convected in the same direction because of electro-osmosis

The  $\text{H}^+$  accumulates outside the exclusion zone because of Donnan exclusion.

# Charge accumulation

According to Maxwell's equations, charge accumulation can only occur over a time period equal to :

$$\frac{\textit{Permittivity}}{\textit{Conductivity}}$$

In the case of the atmosphere this ratio is O(minute)

In the case of water this ratio is O(ns)

# Conclusions

- The optical tweezing experiment measured a viscosity of the EZ- water equal to the viscosity of regular water.
- The particle tracking system measured the movement of beads at the border of the EZ-zone. Within the EZ-zone the beads were moving away from the Nafion. Away from the EZ-zone the beads were moving towards the Nafion.
- Different explanations were explored for the observed vortices: Diffusiophoresis, electrophoresis and electro-osmosis.
- The most plausible explanation was found to be electro-osmosis in a positively charged cristalline scaffold of HBW.

# Acknowledgement

We acknowledge financial support from Framework 7,  
GENODISC grant,  
as well as financial support from Procter and Gamble.

# Future work..

1. Investigating the root cause of the EZ formation and
2. Understanding the confined flow
3. Determining the conditions that leads to crystal formation