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Costa Ribeiro Memorial Lecture
17th B-MRS/SBPMat Meeting

Materials for a Better Future





Synopsis

- A **Golden Age** OR **Decline of Humankind?**
- Biomass: Food, energy and raw materials
 - **Materials from biomass**
- Chemical Electrostatics
- Perspectives

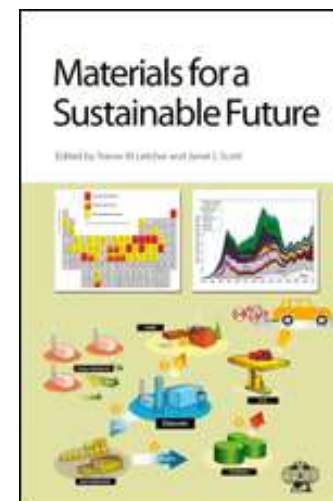
What is ahead?

- Every epoch is marked by **transformation mediated by materials**.
- What are the **upcoming new materials** and how will they transform our lives?
- Will the **needed materials still be available**, in the coming decades?



A Golden Age?

- Prospect: unlimited supply of energy
 - Inexpensive solar energy, competitive wind power, biomass...
 - Fusion, in twenty years (?)
- Inexpensive energy is the key to abundant raw materials
 - e.g. lithium
 - Largest reserves in the world are in: Argentina, Bolivia, Chile, China, US, Australia, Canada.
 - Largest current producers are Australia, Chile, Argentina, China, Zimbabwe...
 - Mg from seawater, Au from “urban mining” ...
 - H₂O from seawater



[Materials for a Sustainable Future](#)
[Scott, J. L. \(ed.\)](#) &
Letcher, T.
(ed.) 2012 Cambridge:
[Royal Society of Chemistry](#). 828
p. *Research output: Book/Report › Book*

In the past: biomass provided food, energy and raw materials

Food: 100%

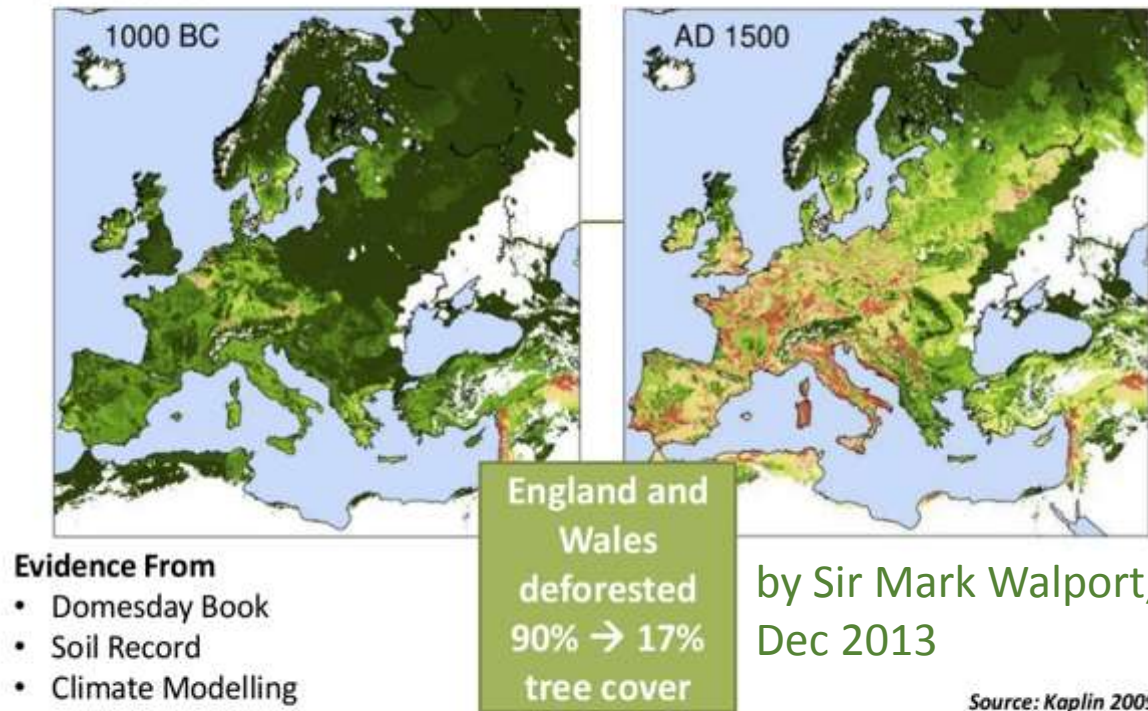
Energy: wood

Raw materials: wood,
natural fibers + minerals

Consequence: land
change

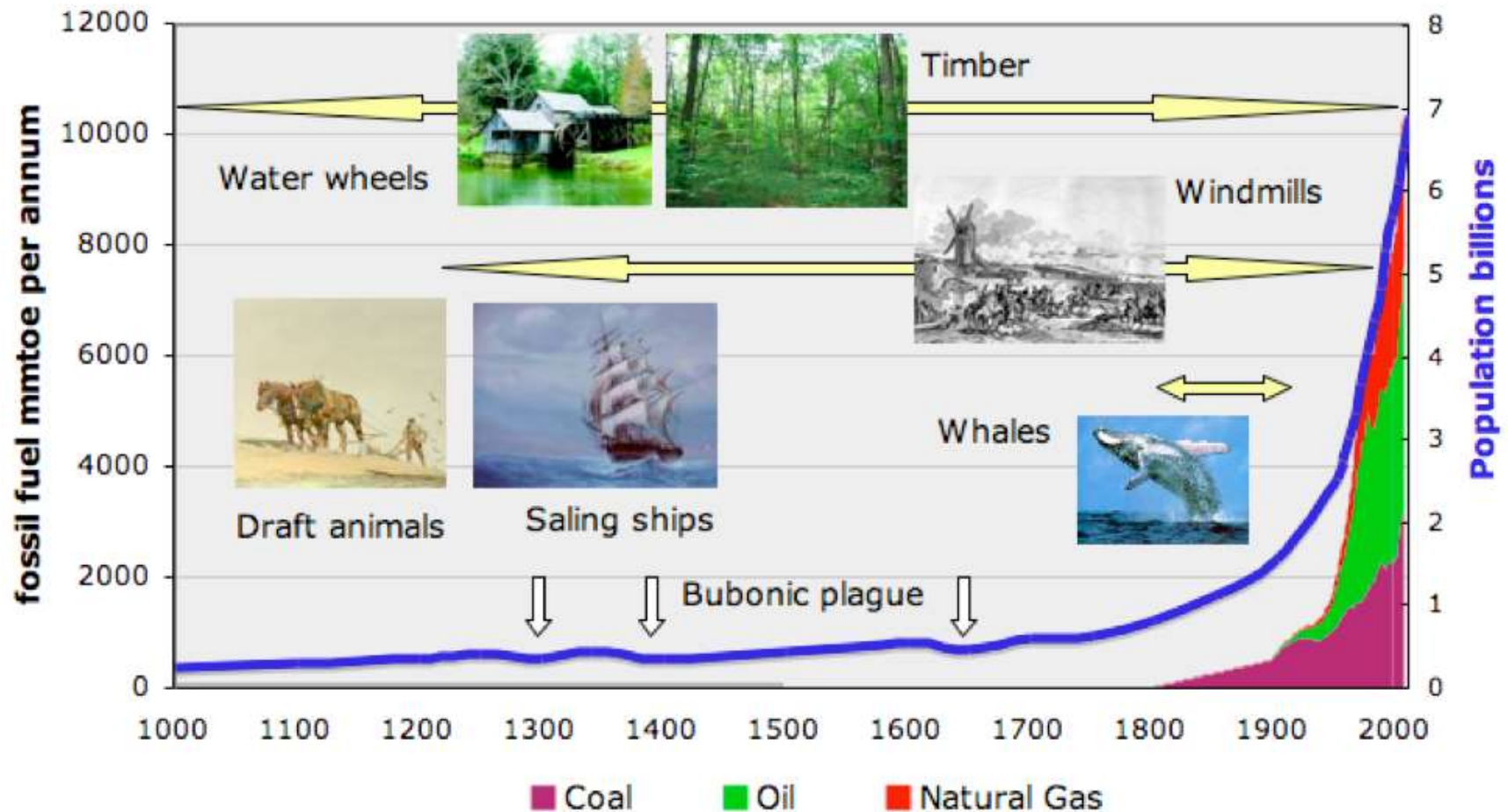

Government
Office for Science

Deforestation of Europe



43 Natural History Museum: Annual Science Lecture, Sharing the Planet

Then came coal, oil and gas



Abundant oil? Make food from oil!

- Use of hydrocarbon fractions for production of single-cell protein
 - Nine plants in the Soviet Union ranged from 50 to 240,000 tons/year.
 - <http://www.nytimes.com/1973/11/10/archives/soviet-plant-to-convert-oil-to-protein-for-feed-use-of-yeast.htm>
- Unesco Science Prize, 1976

France

Alfred Champagnat

BP's Lavera Oil Refinery in France

“pour ses conclusions
sur la production à la
chaîne et à bas coût de
nouveaux proteines
provenant du pétrole”

“for his findings on the
low-cost mass
production of new
proteins from
petroleum”



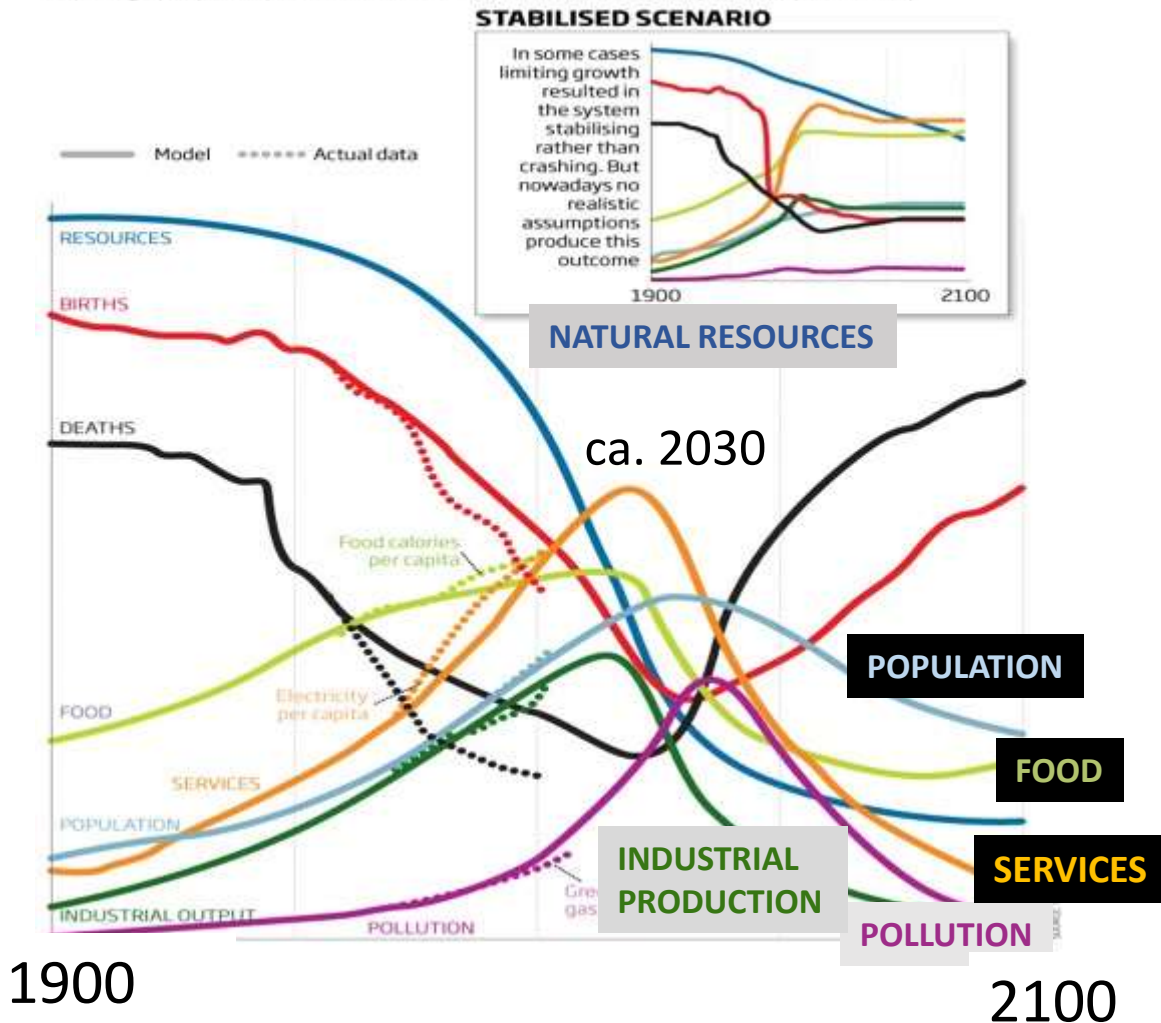
<http://unesdoc.unesco.org/images/0011/001111/111158E.pdf>

Concern: resource depletion. World 3 model

Boom and bust

©NewScientist

In most runs of the World3 computer model, rapid growth is followed by sharp decline. So far the standard run (main graphic) corresponds well with measurements of real-world equivalents (dotted lines)



Solution: energy from renewable resources. Biofuels

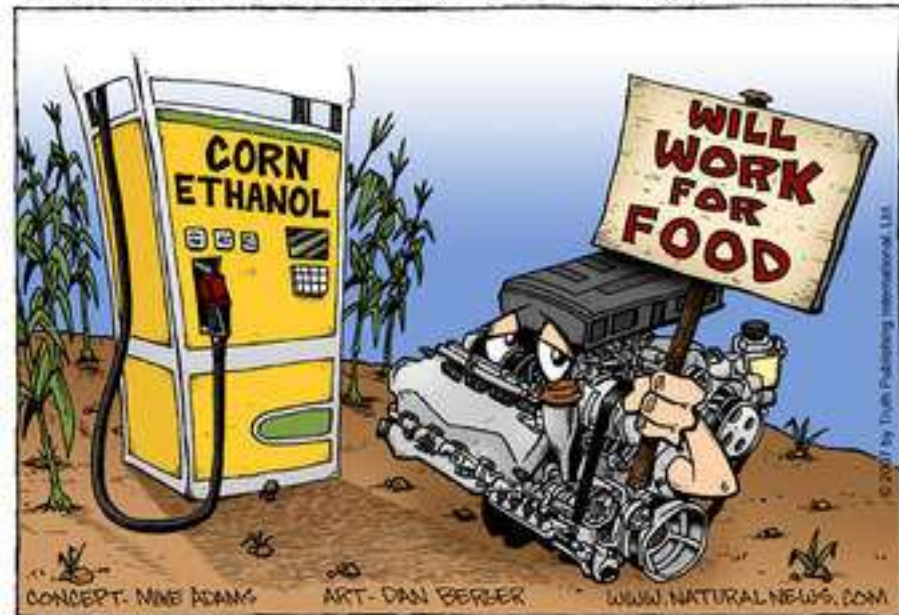


Solution: energy from renewable resources. Biofuels



New Problem

COUNTERTHINK: FUEL VS. FOOD



The Food Crisis (2007)



- The "Food riots"
 - FAO Food Price Index rose to 220 (Jul 2008) and 238(2011)
- Causes:
 - Rising biofuel production from corn, sugarcane, palm oil
 - **Wild commodity speculation, pre 2008 financial crisis**
 - Climate, drought

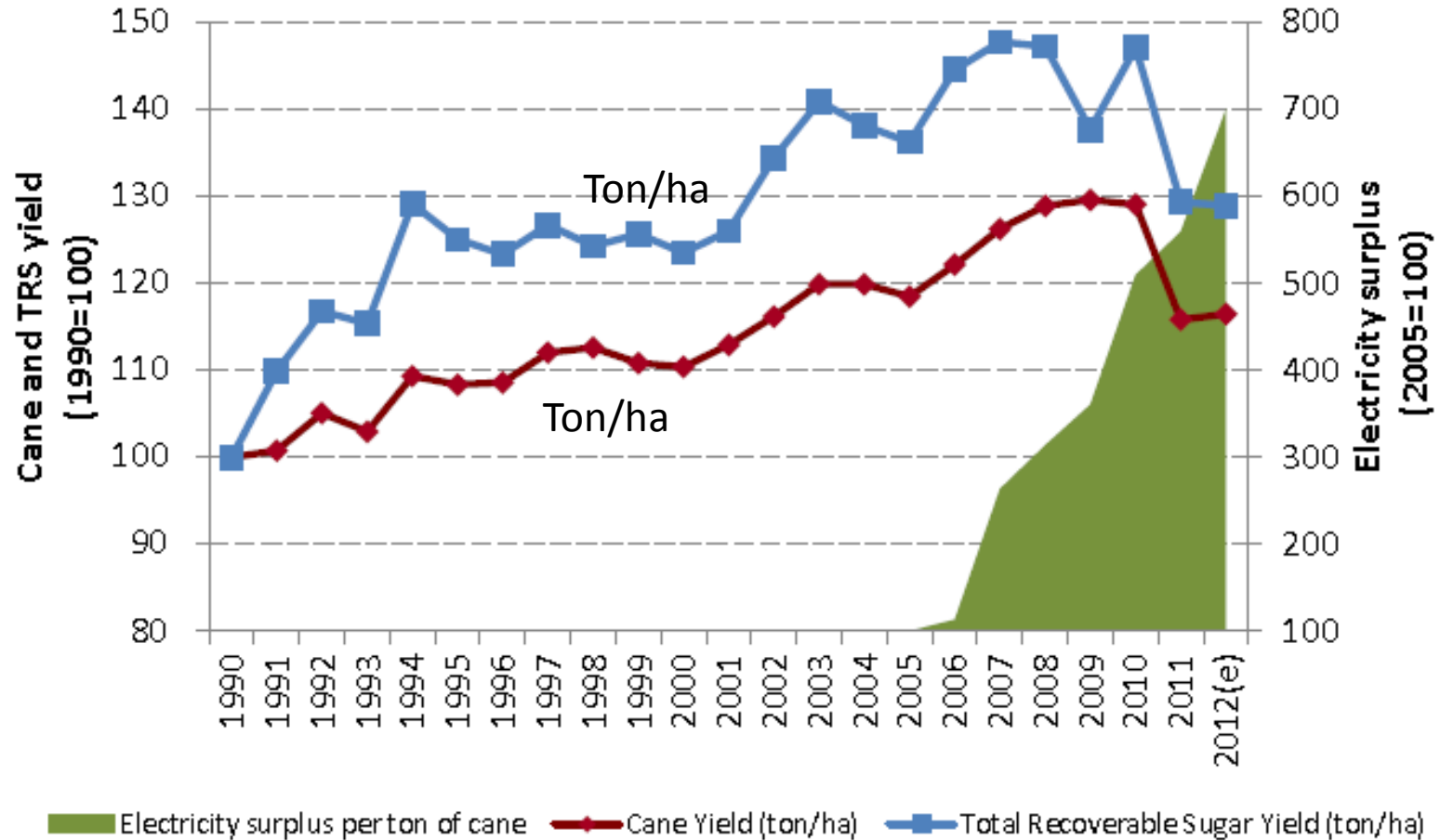
By Jashuah - Own work by uploader, data from Food and Agriculture Organization, CC BY-SA 3.0, <https://commons.wikimedia.org/w/index.php?curid=19394247>

FAO Hunger Map, 2015



<http://www.fao.org/3/a-i4674e.pdf>

Production and efficiency gains: sugarcane



Total recoverable sugar: +39 % from 1990 to 2012.
Electricity surplus: 1 to 11 Twh/year, from 2005 to 2012

Products from sugarcane

- Introduced in South and Central America in the 16th century, to produce sugar.
- Sugar crystallization residues were fermented to produce ethanol.
- In 1976: subsidized fuel ethanol production.
- In 1990: subsidies were eliminated in 90% of the producing areas.
- Sugarcane is the biomass input for making ethanol, butanol, polyethylene, wax, green solvents and surfactants, nanosilica, cellulose, paper and pulp, microcrystalline cellulose, PHB- and other thermoplastics, lysine (>600,000 tons/year), electricity.
- Galembeck, Csordas “Chemicals from Sugarcane”, in Fletcher and Scott “Materials for a Better Future”, RSC 2012



Biofuels **contribute** to food security

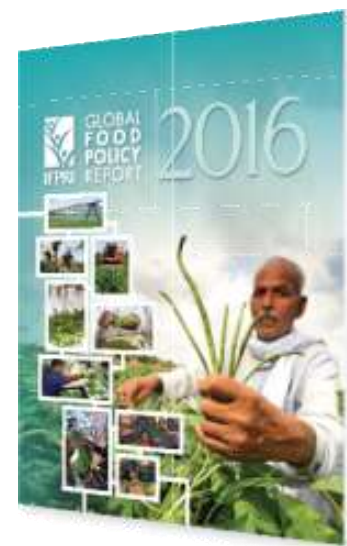
“...biofuels may offer an opportunity to improve food security.

Yet, for many poorer countries in Africa and elsewhere, biofuels may be better viewed as a potential export or as a means for reducing fossil fuel imports.

...producing conventional biofuels in low-income countries could raise rural incomes beyond what is required to offset rising food prices.

Studies in Ethiopia: ... **farmers' participation in biofuel programs** encouraged greater use of fertilizers and improved farming technologies, **leading to higher food-crop productivity and better food security during the year.**

One precondition for success, however, was farmers' access to high-quality, productive biofuel crops.”



C. Arndt et al., Fueling the path to food security, IFPRI Global Food Policy Report 2016 p.63

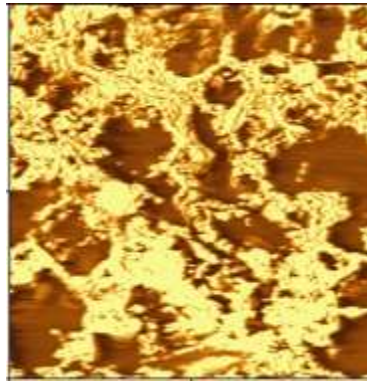
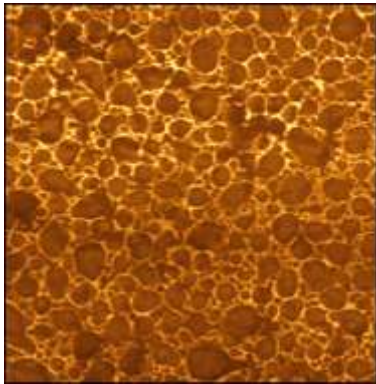
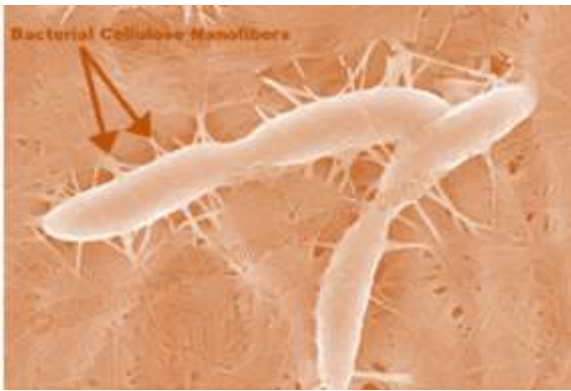
F. Galembeck, Synergy in food, energy and advanced materials production from biomass.

Energy and Environmental Science 2010 and Pure and Applied Chemistry 2018.

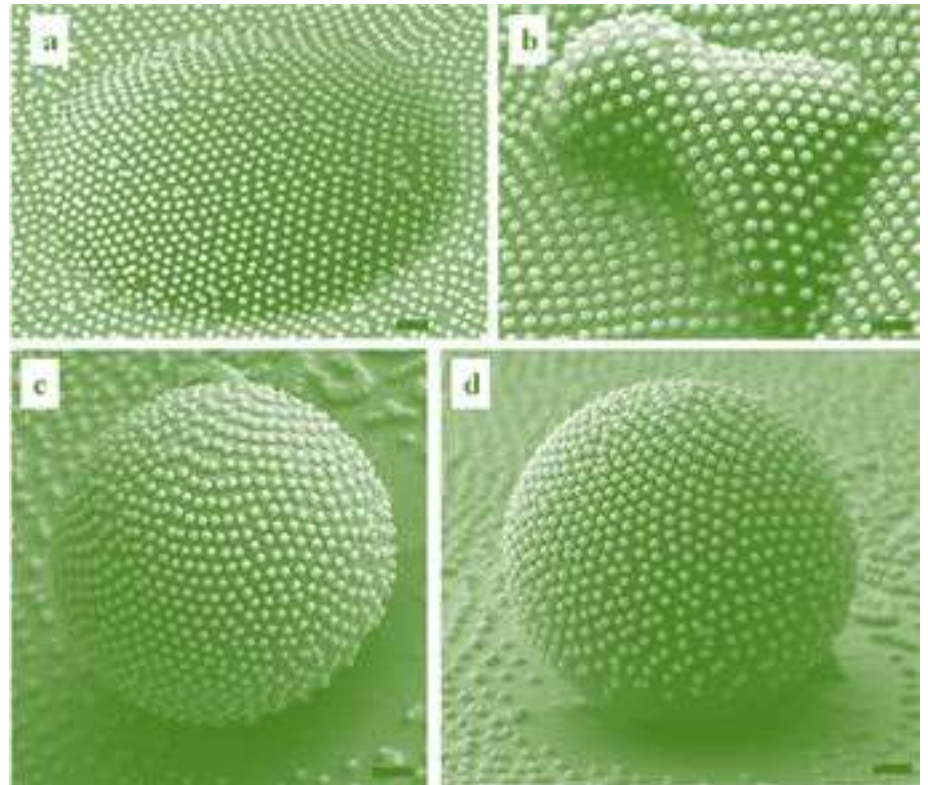
A note on policy

- What do policymakers expect from scientists?
 - Not much.
 - Few citations in “The UN Sustainable Development Goals Report” 2018. “science” 3 times; “scientific”: 1; “research”: 2; “technology/ical/ies”: 21.
- Brazilian science concentrates on explaining, modelling, theorizing...
 - Much smaller effort on making.
- Result: small number of significant new products and processes,
 - ...although all the competitive economic sectors rely heavily on S&T.

Materials from Biomass: Well-known or Ever-surprising?



- Natural Rubber
 - Cellulose
- Electrostatic Adhesion

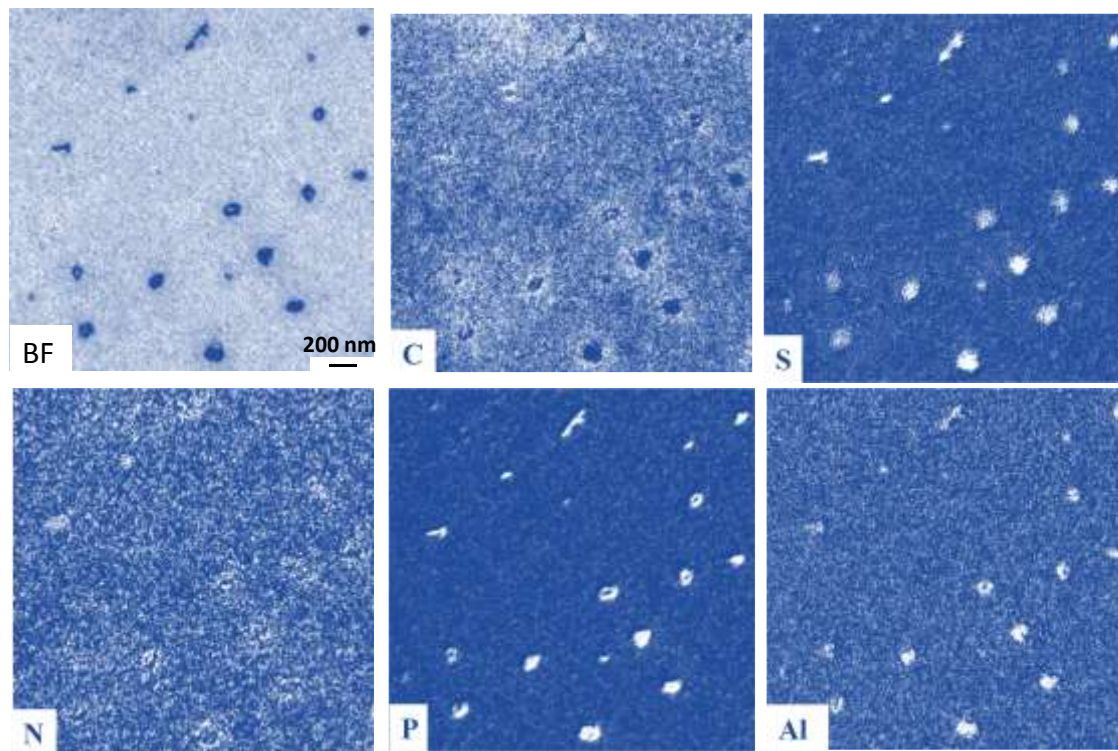


Natural Rubber



- Accounts for 42% of global elastomer consumption
- Essential in transportation and many industrial products Rubber Statistical Bulletin April-June edition, quoted by S. Rolere, et al. in European Polymer Journal (2016)
- The “mystery of natural rubber”
 - “...as the tire size increases and the level of punishment the tire will take goes up, the amount of natural rubber increases...” (Josh Velson)
- Which is the role of the non-rubber constituents (inorganics, proteins and phospholipids NR properties?)

ESI-TEM thin film elemental maps of dialyzed natural rubber latex film

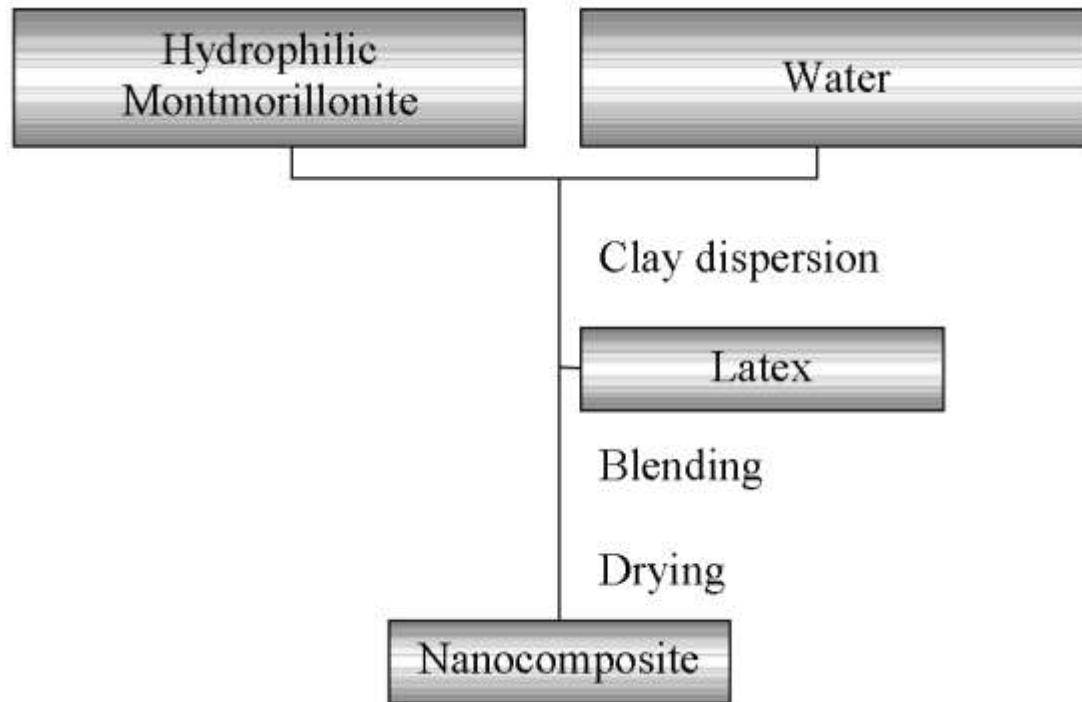


- **Adherent mineral nanoparticles** in natural rubber
- C, N densification around the inorganic particles: **adhesion**

Rippel, M.M. et al. *Analytical Chemistry*, 2002

Learning from Nature

Preparation of nanocomposites in aqueous media using latex



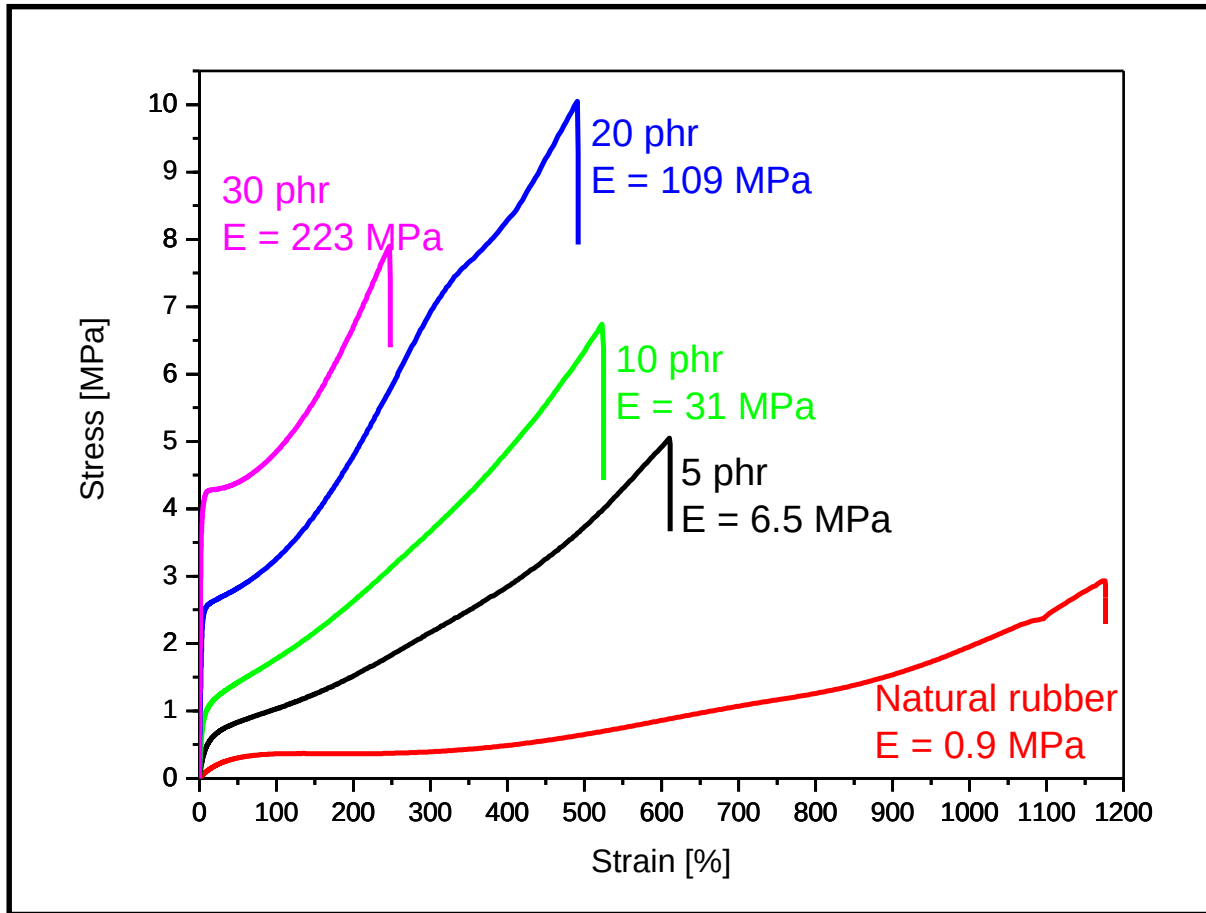
Varghese, S., Karger-Kocsis, J., ***Polymer***, **2003**, 44, 4921.

Wu, Y.P., Wang, Y.Q., Zhang H.F. *et al.* ***J. Compos. Sci. Tech.*** **2005**, 65, 1195

Valadares, L. F., Leite, C. A. P., Galembeck, F., ***Polymer***, **2006**, 47, 672.

Valadares, L. F., Murakami, M. M., Rippel, M. M., Galembeck, F., PI0301193-3, **INPI**, **2003**.

Tuning-up mechanical properties



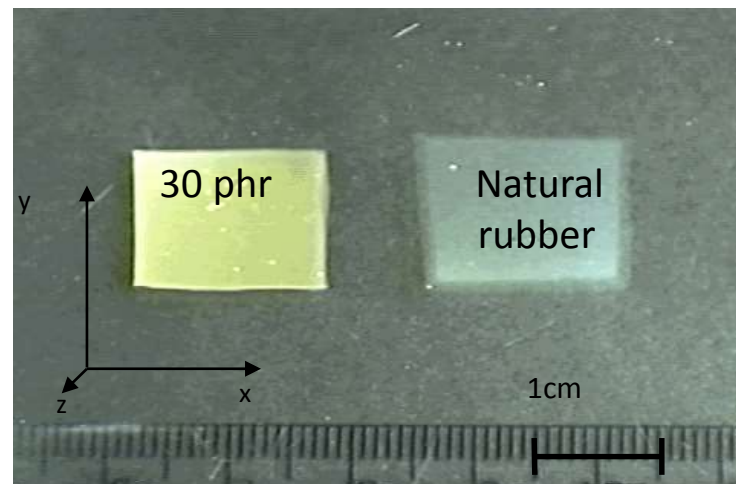
Tensile testing:
Modulus increases
ca. 250 times
Maximum stress
increases up to 3.5
times

Valadares, L. F., Leite, C. A. P., Galembeck, F., *Polymer*, 2006, 47, 672.

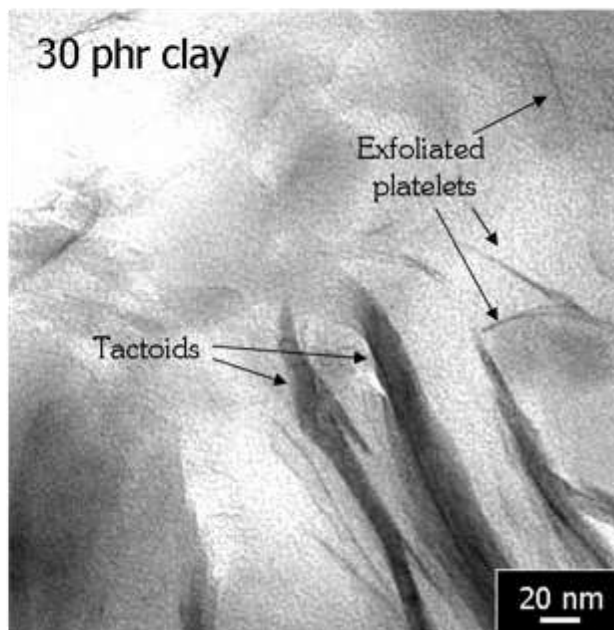
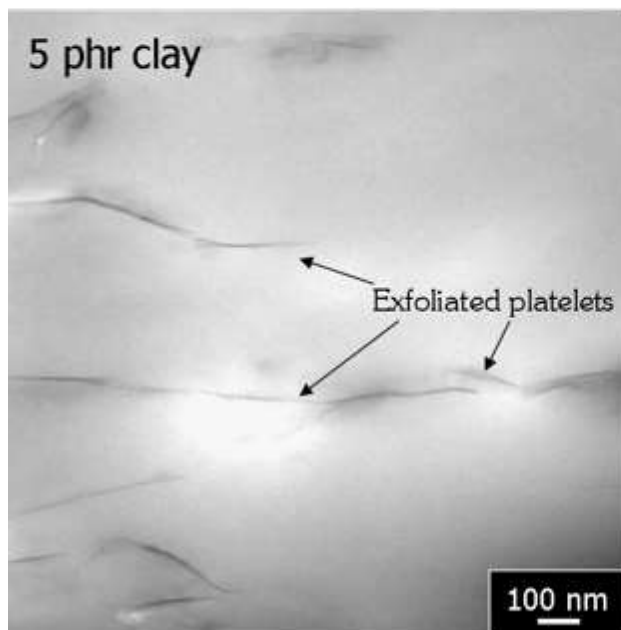
Strong polymer – clay adhesion

Formation of a polymer-particle
network

Resistance to solvents



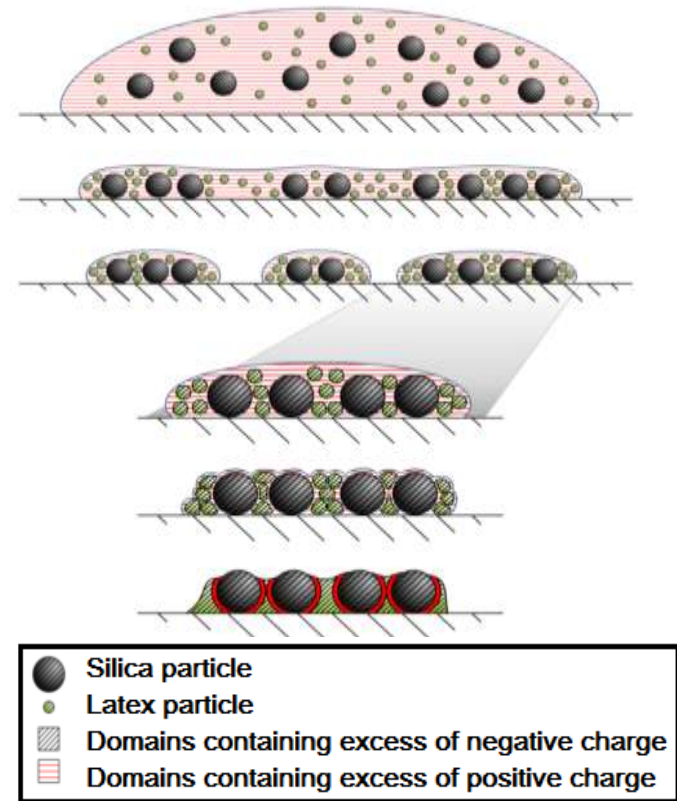
Transmission Electron Microscopy



Valadares, L. F., Leite, C. A. P., Galembeck, F., *Polymer*, 2006, 47, 672.

Composite formation and stability

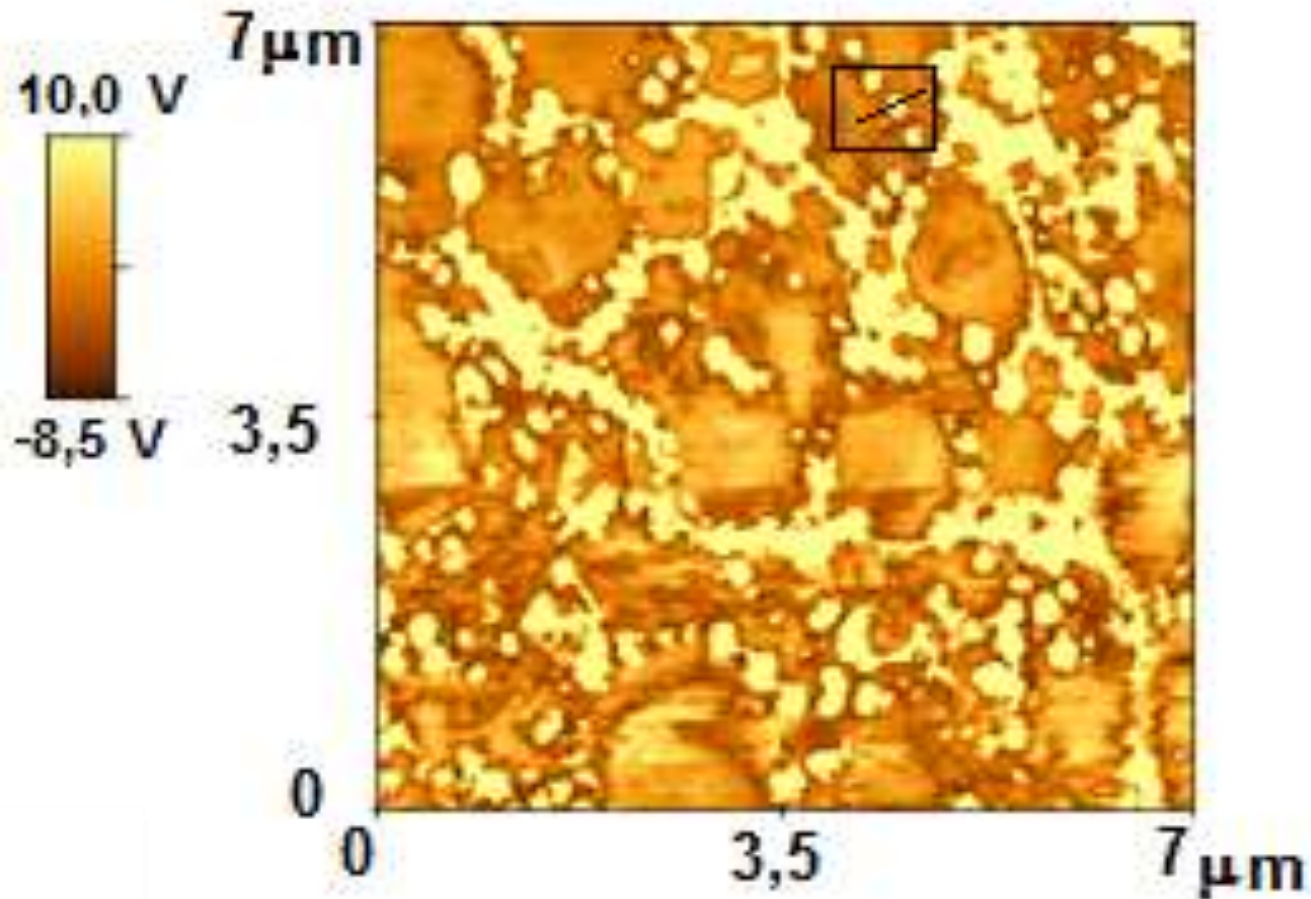
- Capillary adhesion during drying
- Electrostatic adhesion in the dry monolith
- Both are controlled by interfacial properties
- Water-based processes for making composites from incompatible materials



Valadares, L.F et al.. *J. Phys Chem C* (2008); Bragença, F. C. ; et al. *Chemistry of Materials*, (2007)

Kelvin micrograph from PVC / natural rubber blend

Electrostatic contribution to cohesion



Linares et al., Langmuir 2008

Food waste



Nanocarbon dispersion



Natural rubber



NC/NR
composite



Overcoming
limits to
miscibility:
conductive
“green” rubber

Kampioti, Matos
et al.,
ACS Omega 2018

Conductive inks, paints and adhesives

- Usually made of silver
 - Also: copper, PEDOT, graphite, carbon nanotubes, graphene.
- US\$ 3 Billion/year market
 - CAGR = 4 – 6 %, depending on the market or product segment
- Broad range of users
 - Auto industry: window and seat heaters
 - Hardware and displays: screens, contacts
 - Touch screens: ITO, graphene, carbon nanotubes
- Rao R, V. K., Abhinav K, V. et al. (2015).Conductive silver inks and their applications in printed and flexible electronics. RSC Adv. 5, 77760–60.
- Yang, W. and Wang, C. (2016) Graphene and the related conductive inks for flexible electronics. J. Mater. Chem. C 4, 7193– 207.

THE MAIN PLAYERS

Material	Resistivity ($\Omega\cdot\text{m}$)	Resistance (Ω/sq)	Thickness (m)
graphene <i>monolayer</i>	10^{-8}	33	$3 \cdot 10^{-10}$ (single sheet)
graphene <i>nanosheet</i>	10^{-6}	33 (1)	$3 \cdot 10^{-8}$ (100 sheets)
graphite (basal)	$2 \cdot 10^{-6}$	0.3	$6 \cdot 10^{-6}$ (20K sheets)
graphite (microfluidized)	2.5×10^{-4}	2 (2)	
silver	1×10^{-8}	10^{-3}	10^{-5}
silver ink	12.5×10^{-8}	5×10^{-3}	2.5×10^{-5}

(1) Fang, X. Y., Yu, X. X. et al. (2015) Temperature and thickness-dependent electrical conductivity of few-layer graphene and graphene nanosheets, *Physics Letters A*, 379, 2245-51.

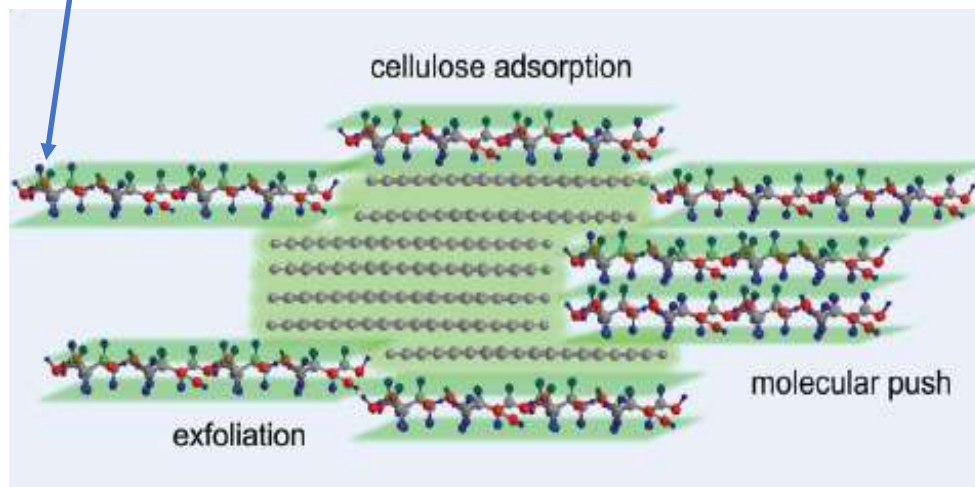
(2) P. G. Karagiannidis, S. A. Hodge, L. Lombardi et al., Microfluidization of Graphite and Formulation of Graphene-Based Conductive Inks, *ACS Nano* 2017, 11, 2742-2755.

Cellulose is soluble in water + alkali

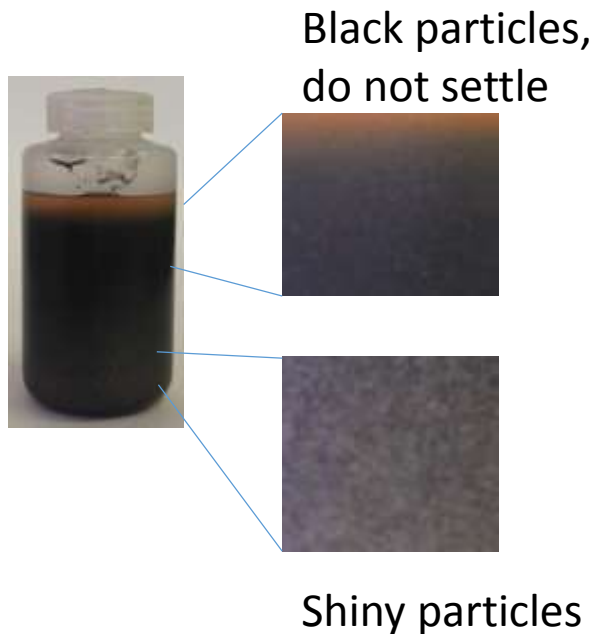
- Simple procedure: Isogai et al. *Cellulose*, **5** (1998) 309
- Insolubility in pure liquids derives from its amphiphilic structure (the Lindman hypothesis)
- Shows the usual properties of polyelectrolytes. Adhesive for paper, cloth, wood.
 - E. S. Ferreira et al, *ACS Applied Materials & Interfaces* (2015)

- Excellent dispersant and exfoliating agent for graphite.

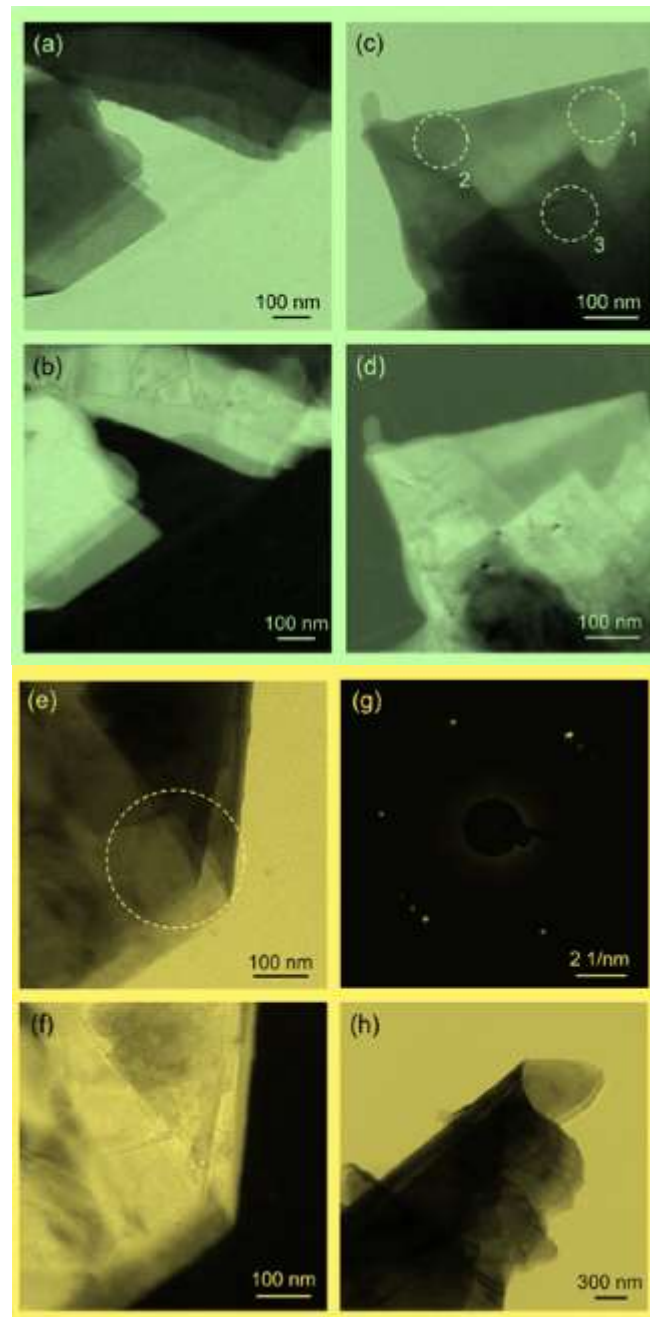
- E. S. Ferreira et al, *Nanoscale* (2017)



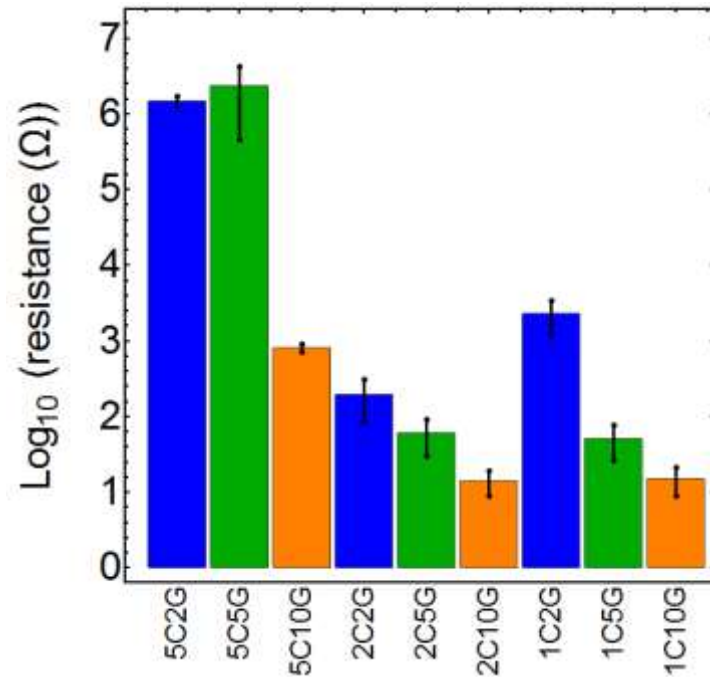
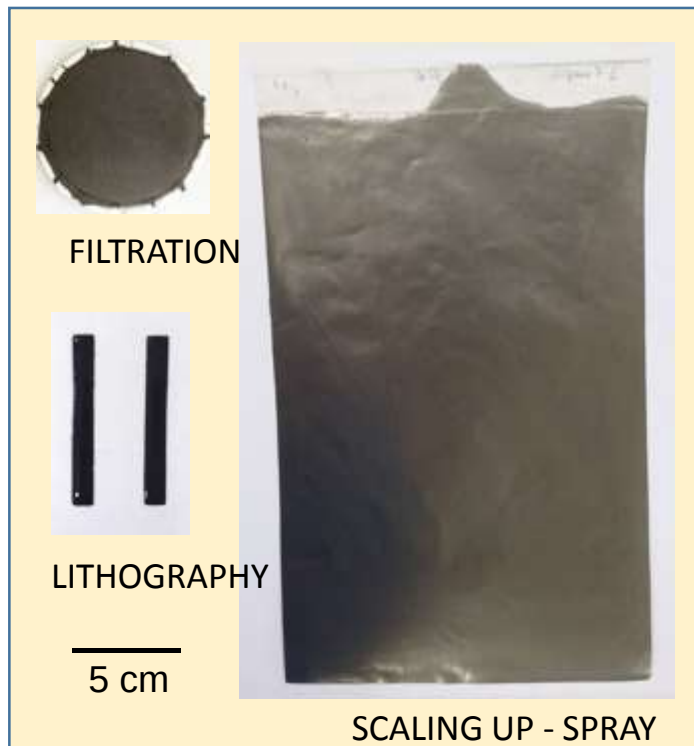
Graphite **exfoliated** in alkaline aqueous cellulose solution



E. S. Ferreira et al, *Nanoscale* (2017)



Graphite coatings on paper, cloth and ceramics



Broad resistance range
depending on cellulose/graphite
ratio

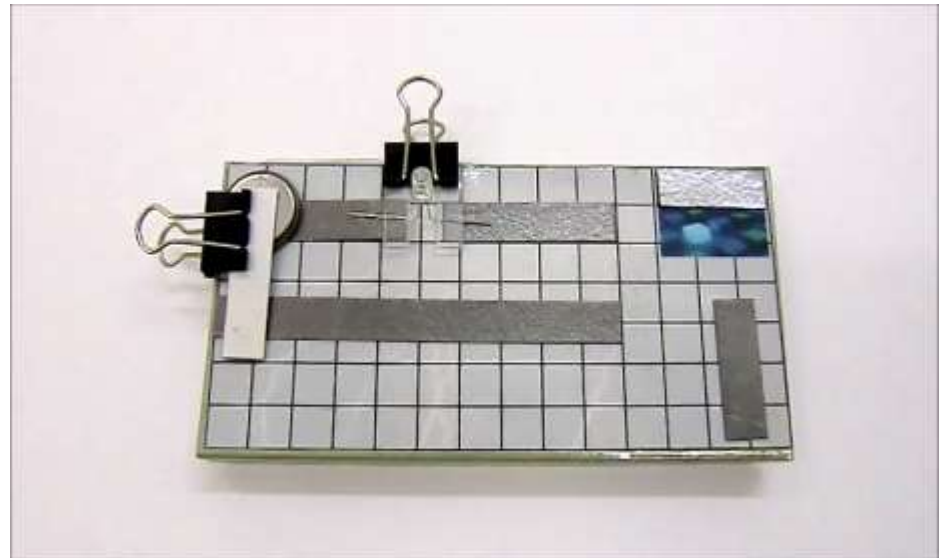
Typical thickness: 10 – 100 μm

Benchmarking



Material	Resistivity ($\Omega\cdot\text{m}$)	Resistance (Ω/sq)	Thickness (m)
graphene <i>monolayer</i>	10^{-8}	33	$3 \cdot 10^{-10}$ (single sheet)
multilayer graphene	10^{-6}	33	$3 \cdot 10^{-8}$ (100 sheets)
graphite (basal)	$2 \cdot 10^{-6}$	0.3	$6 \cdot 10^{-6}$ (20 K sheets)
graphite/cellulose	$3.7 \cdot 10^{-5}$	0.3	$125 \cdot 10^{-6}$ (total)
microfluidized graphite	$2.5 \cdot 10^{-4}$	2	
commercial silver ink	$12.5 \cdot 10^{-8}$	$5 \cdot 10^{-3}$	$2.5 \cdot 10^{-5}$

Inexpensive, safe circuit boards...



...electrodes, sensors,
supercapacitors, heaters...

Electrostatics and Friction: Fundamental but Snubbed

Well-established knowledge

OR

Unsolved problems?

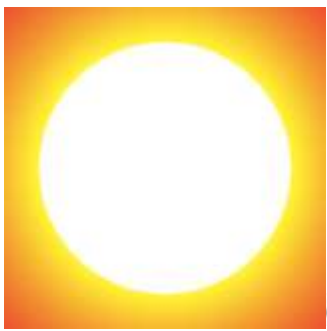
Neglected as scientific topics in the 20th century

Disconnected from atomic-molecular theory

Revived interest since the first decade of this century

Electrostatics is Alive, Again

- We live in an electrified environment
- Electricity is produced within the Earth capacitor
- Electroneutrality is the exception, anywhere
 - The charge carriers
 - Chemical electrostatics
- The atmosphere is a charge reservoir
 - Fires, explosions, safety
- Outlook: new science, new technology



Solar energetic protons

An electrified environment: the Global Electric Circuit

Field-aligned currents

Auroral currents

Geomagnetically
induced currents

Fair weather
return
currents

Transient
luminous events

Thunder
Clouds

Magnetic
Field Lines

Electrified
Clouds

Galactic Cosmic Rays

>100 km
Upper

100 km
Middle

10 – 15 km
Lower

Electric potential gradients at Kew Observatory, London, 1966 to 1979):

$50 - 1200 \text{ V m}^{-1}$

Rycroft and Harrison, Space Sci Rev 2008

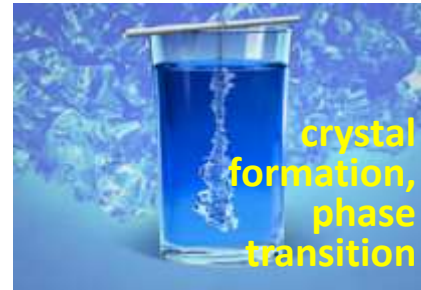
How electricity is produced, in the atmosphere?

“...current geophysical research has not yet disclosed effective models for atmospheric cloud electrification.”

Helsdon Jr., J. H.; Gattaleeradapan, S., Farley, R. D.; Waits, C. C.
J. Geophys. Res. **2002**, 107, 4630.



water splash



crystal
formation,
phase
transition



friction and wear



aerosol

We add electricity to the environment
(beyond power plants and the grid)

Charge separation/ interfacial effects:
Maxwell-Wagner-Sillars

Costa Ribeiro

Workman-Reynolds



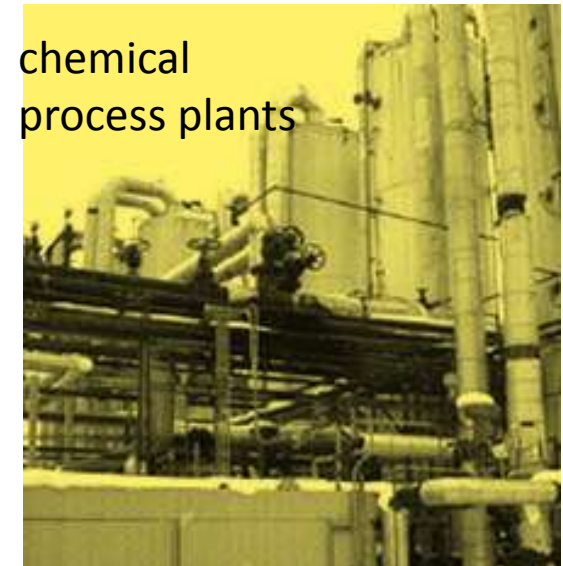
milling



powder
handling

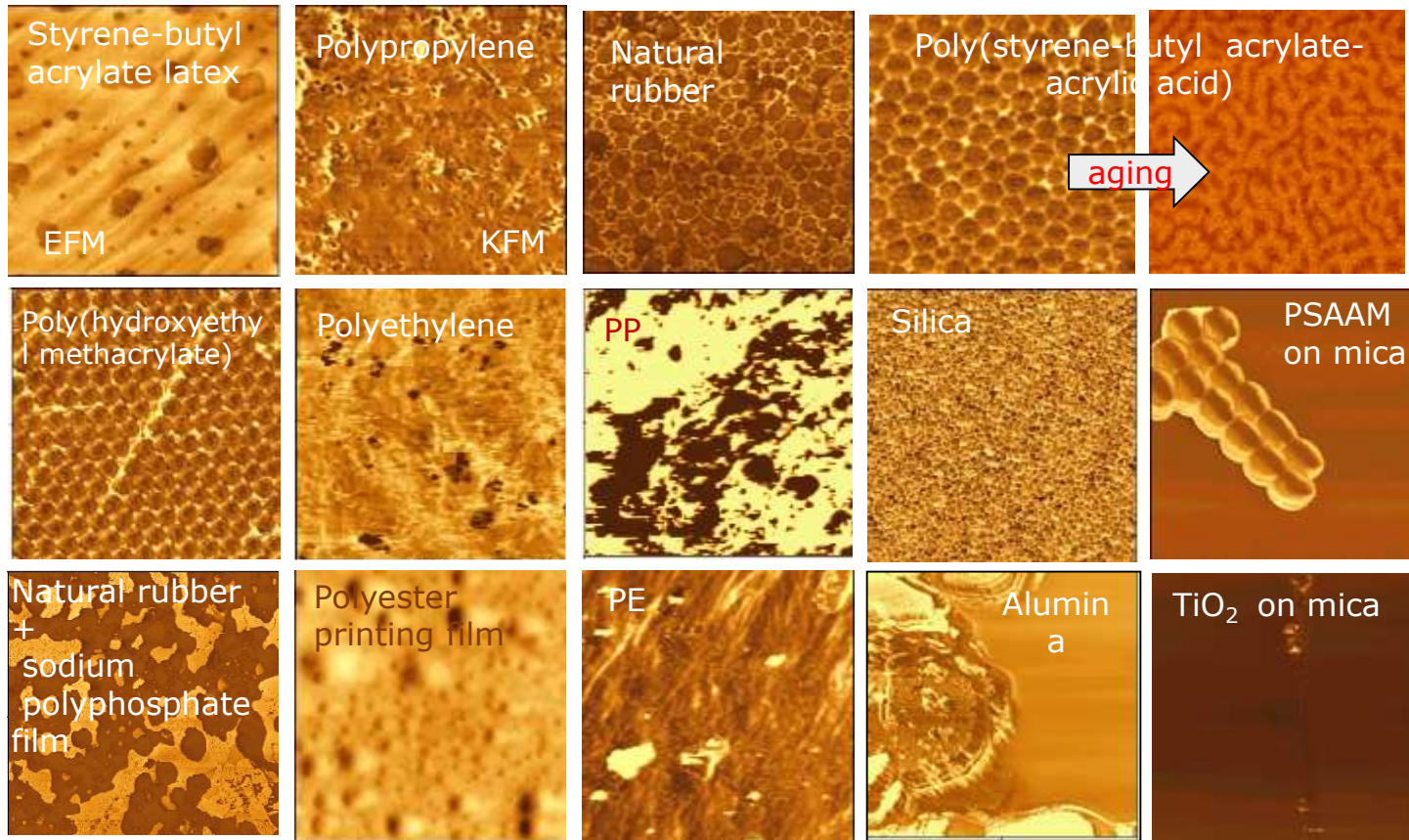


R
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chemical
process plants

Unexpected: Complex charge distribution in dielectric solids

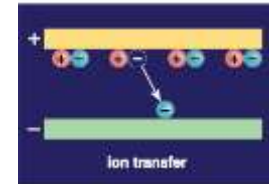


A Galembeck, CAR Costa, et al., *Polymer*, 2001, 42, 4845; F Galembeck, CAR Costa, et al., *An. Acad. Bras. Cienc.*, 2001, 73, 495; FG, C Rezende, RF Gouveia, MA da Silva *Journal of Physics – Condensed Matter*, 21, 2009, 26300

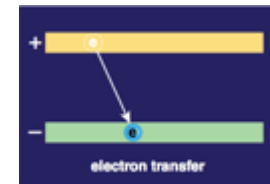
Why? Water ion partition/adsorption and mechanochemical reactions

Charge carriers are electrons or ions?

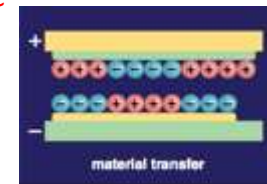
Ions: McCarty and Whitesides, *Electrostatic Charging due to Separation of Ions at Interfaces: Contact Electrification of Ionic Electrets*, *Angew. Chem. Int. Ed.* 2008, 47, 2188-2207. **Ions**, Baytekin, Grzybowski, et al., *The Mosaic of Surface Charge in Contact Electrification*, *Science* 2011, 333, 308-312.



Electrons: Liu and Bard, *Electrostatic electrochemistry at insulators*, *Nat. Mater.* 2008, 7, 505-509.



Ions, Mass Transfer: Burgo, Galembeck et al., *Triboelectricity: Macroscopic charge patterns formed by self-arraying ions on polymer surfaces*. *Langmuir* 2012.



Critical analysis: Meurig W. Williams (2012)
American Scientist 2012, 100(4), 316-325.

Feature Article: **What Creates Static Electricity?**

“Traditionally considered a physics problem, the answer is beginning to emerge from chemistry and other sciences.”

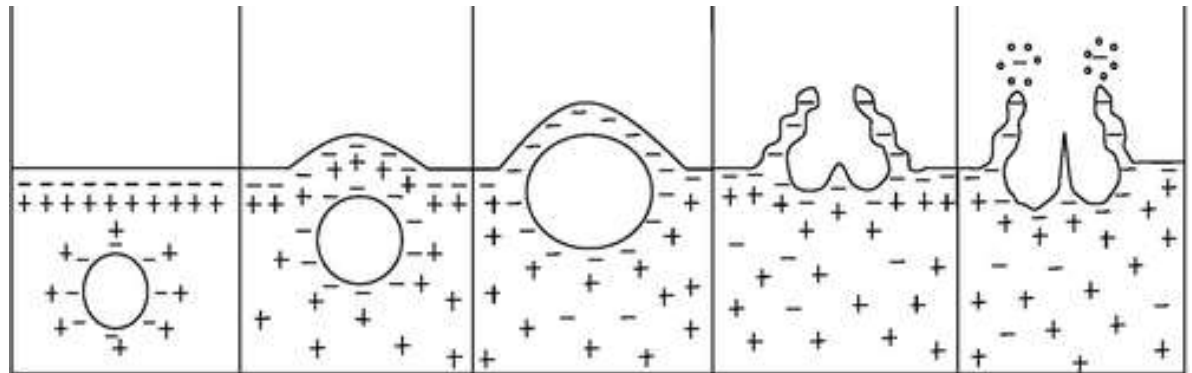
Electroneutrality: exception, not the rule



- “Electroneutrality principle: The principle expresses the fact that all pure substances carry a net charge of zero.” (IUPAC Gold Book)
 - Can only happen under **zero electric potential**: $\mu_- = \mu_-^\circ + RT (\ln x_-) + zFV$
 - Pauling’s Electroneutrality Principle **of molecular structure**: **unduly extrapolated** to macroscopic matter in introductory science courses.
- Charge partition and accumulation are expected in any **interface**: **Maxwell-Wagner-Sillars effect**.
- Charge patterns are **observed in any material system**.
- **The discovery tools**: Kelvin electrode, Kelvin force microscopy and other techniques derived from AFM.

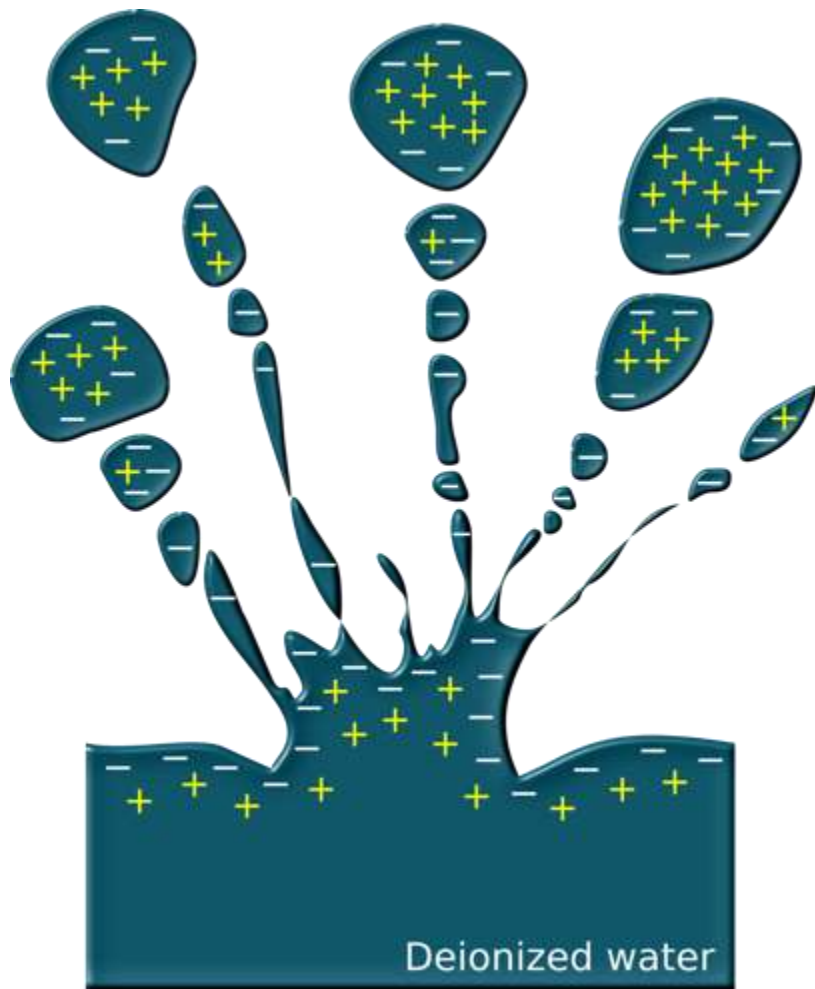
Is water
electroneutral?
Seldom.

- Ovchinnikova, K.; Pollack, G. H. *Langmuir* 2009, 25, 542: **Can water store charge?**
- Corti, H. R.; Colussi, A. J. *Langmuir* 2009, 25, 6587: criticism to Pollack.
- Amin, M. S.; Peterson, T. F.; Zahn, M. *J. Electrostatics* 2006, 64, 424: **water from different sources carries excess negative charge.**
- McCarty, L. S.; Whitesides, G. M. *Angew. Chem. Int. Ed.* 2008, 47, 2188: **non-electroneutrality should be considered.**
- I. Bhattacharyya, J. T. Maze, G. E. Ewing,* and M. F. Jarrold*, *J. Phys. Chem. A*, 2011, 115, 5723: **bursting droplets are negative.**



Water and electricity

- **Steam electricity**
 - Discovered in 1840 by Lord Armstrong: **vapor is charged opposite to the remaining liquid.**
 - M. Faraday, *Phil. Trans.* 1843, 133, 17.
- **Schrödinger** (Ph.D. Thesis)
 - Passive role for water in static electricity dissipation
- Water ion partition at interfaces
 - **Gas bubbles and pure hydrocarbon drops** in water hold negative charge
 - Beattie, J. K. *Angew. Chem. Int. Ed.* 2004, 43, 3568. Marinova, K. G. *Langmuir* 1996, 12, 2045. **Healy, T. W.**; Fuerstenau, D. W. *J. Colloid Interface Sci.* 2007, 309, 181. I. Bhattacharyya et al., *J. Phys. Chem. A*, 2011, 115, 5723.
 - **Kelvin dropper**
 - <http://ocw.mit.edu/...demonstration: kelvin water dropper>

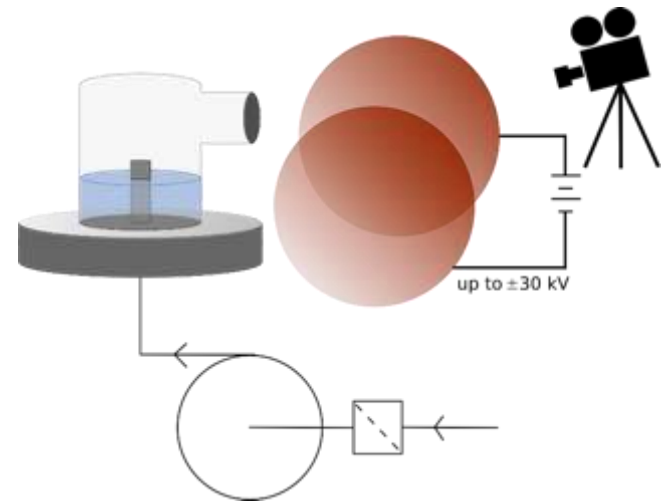


Charge separation due to H₂O partition at interfaces

- **expansion of wet steam:**
 - J. Finke, J. Electrostat. 1989, 23, 69.
- **adsorption-desorption:**
 - R. F. Gouveia and F. Galembeck, J. Am. Chem. Soc. 2005; J. Phys. Chem C 2005, 2008; Anal. Chem. 2011.
- **water ion partition:**
 - T. A. L. Burgo, F. Galembeck, G. H. Pollack, J. Electrostat. 2016, 80, 30–33; T. R. D. Ducati, L. H. Simões, F. Galembeck, Langmuir 2010, 26, 13763–13766; T. A. L. Burgo, F. Galembeck, J. Braz. Chem. Soc. 2016, 27, 229–238; T. A. L. Burgo, T. T. D. Ducati, K. R. Francisco, K. J. Clinckspoor, F. Galembeck, S. E. Galembeck, Langmuir 2012, 28, 7407–7416
- **splashing and bubble breaking:**
 - P. Lenard, Ann. Phys. 46, 584, 1892; Z. Levin, P. V. Hobbs, Phil. Trans. 269, 555, 1971. P. Kolarž et al., Atmos. Chem. Phys. 2012, 12, 3687. I. Bhattacharyya et al., J. Phys. Chem. A 2011, 115, 5723. T. L. Burgo et al., Coll. Interf. Sci. Comm. 2015, 7, 7-11

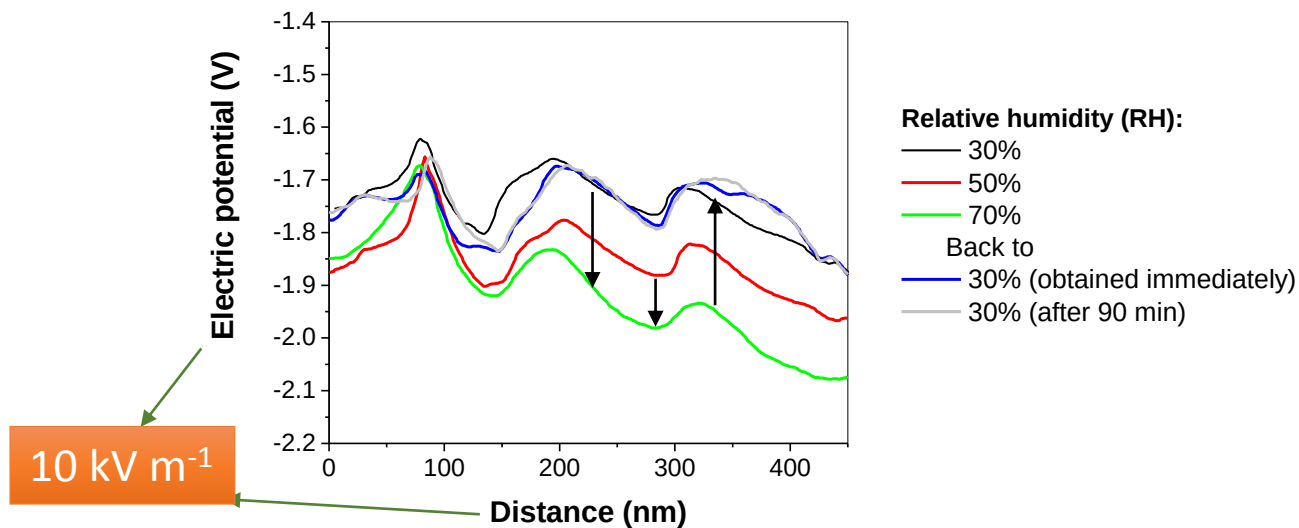
Charge on Aerosol

Positive + negative droplets



Aerosol	Collected liquid weight (g)	
	Electrode (+)	Electrode (-)
Deionized water	0.053± 0.025	0.139 ±0.040
NaCl solution	0.205± 0.066	0.085± 0.017

Water vapor adsorption contributes charge to solids



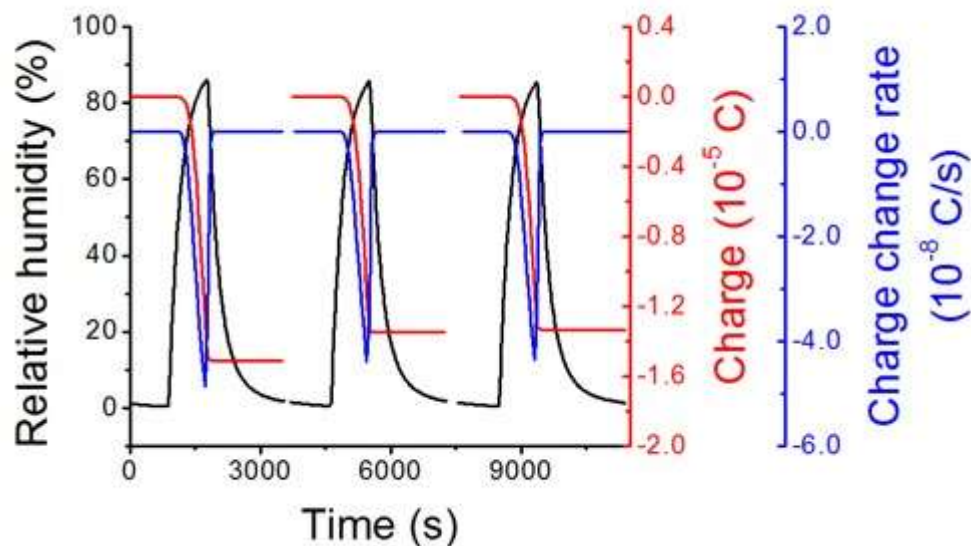
Stöber silica is more negative under high humidity. Reversible.

Atmosphere is a charge reservoir for insulators.

Gouveia, R. F.; Galembeck, F. *J. Am. Chem. Soc.* 131(32), 2009, 11381

Water vapor adsorption contributes charge to isolated metals

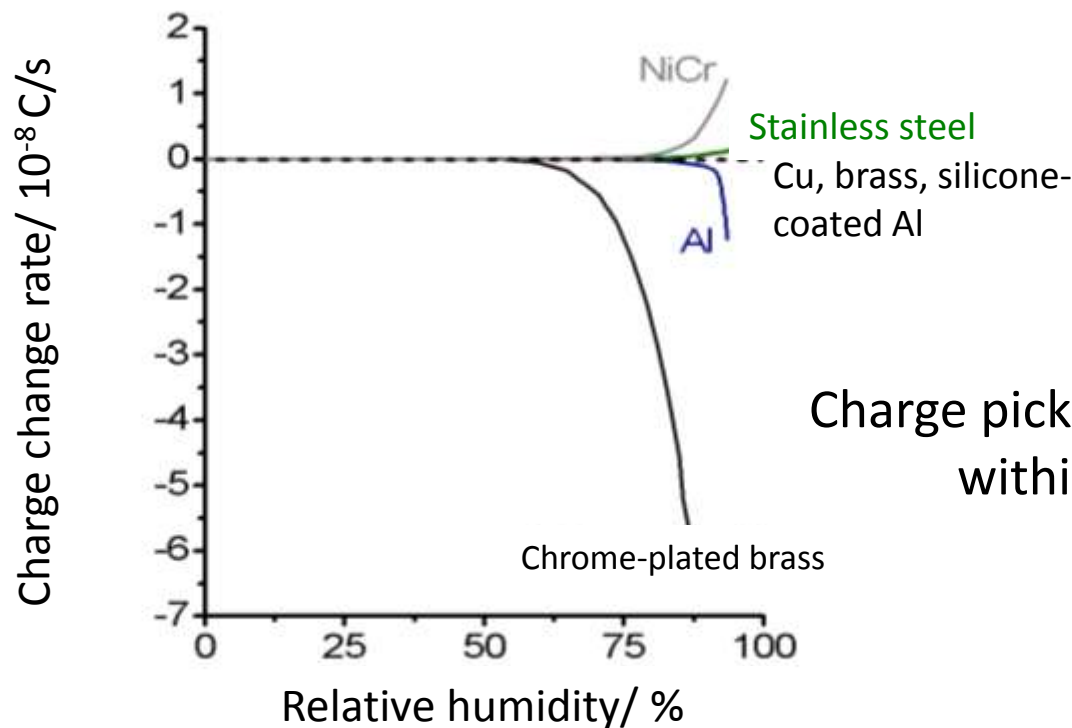
Chrome-plated brass cylinders acquire negative charge under high humidity



“Charge Partitioning at Gas-Solid Interfaces: Humidity Causes Electricity Buildup on Metals”

Telma R. D. Ducati, Luís H. Simões, Fernando Galembeck. *Langmuir*, **2010**, 26, 13763-13766.

Water vapor adsorption contributes charge to isolated metals



Charge pick-up by **isolated metals** within a Faraday cup

"Charge Partitioning at Gas-Solid Interfaces: Humidity Causes Electricity Buildup on Metals"

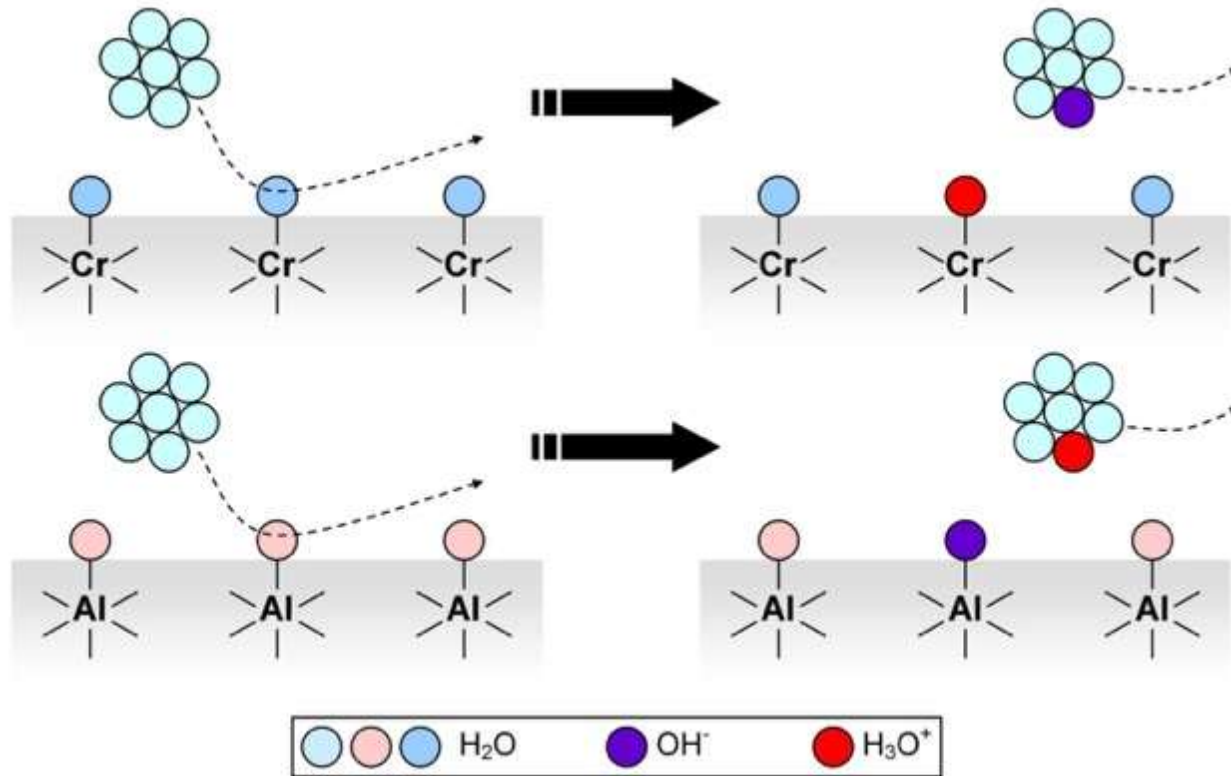
Telma R. D. Ducati, Luís H. Simões, Fernando Galembeck. *Langmuir*, **2010**, 26, 13763-13766.

Acidic surfaces become more negative when the humidity changes from 30 to 70%

Basic surfaces become more positive.

Substance	ΔV_m	$(\Delta V)^2_m$	$(\Delta V_m)^2/(\Delta V)^2_m$	
Iron oxide@Fe	-0.464 ± 0.004	0.217 ± 0.004	0.99	Acid
Magnesium sulfate	-0.229 ± 0.008	0.063 ± 0.009	0.83	Acid
Silica	-0.172 ± 0.015	0.033 ± 0.005	0.90	Acid
Cellulose	-0.104 ± 0.007	0.011 ± 0.001	0.98	Acid
Aluminum oxide@Al	-0.055 ± 0.016	0.004 ± 0.002	0.76	Acid
Calcium oxide	+1.657 ± 0.11	2.811 ± 0.33	0.98	Base
MgO (30- 50% RH)	+0.195 ± 0.062	0.096 ± 0.021	0.40	Base
Nickel oxide	+0.060 ± 0.017	0.005 ± 0.002	0.72	Base
Aluminum phosphate	+0.039 ± 0.007	0.002 ± 0.001	0.76	Base

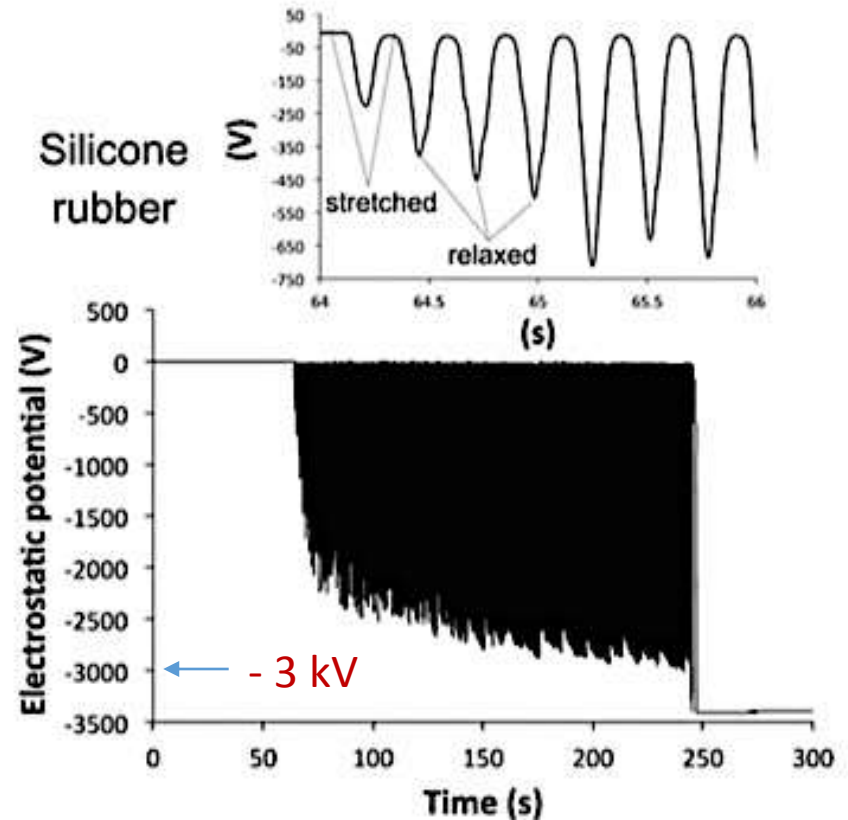
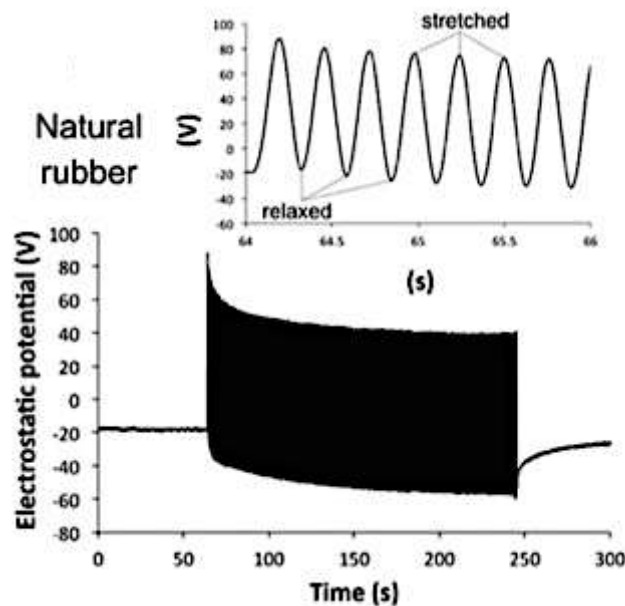
Charge transfer during H₂O adsorption



"Charge Partitioning at Gas-Solid Interfaces: Humidity Causes Electricity Buildup on Metals" Telma R. D. Ducati, Luís H. Simões, Fernando Galembeck. *Langmuir*, **2010**, 26, 13763-13766.

Elastoelectricity

Periodically stretched rubber tubing shows **potential variation in phase** with length.

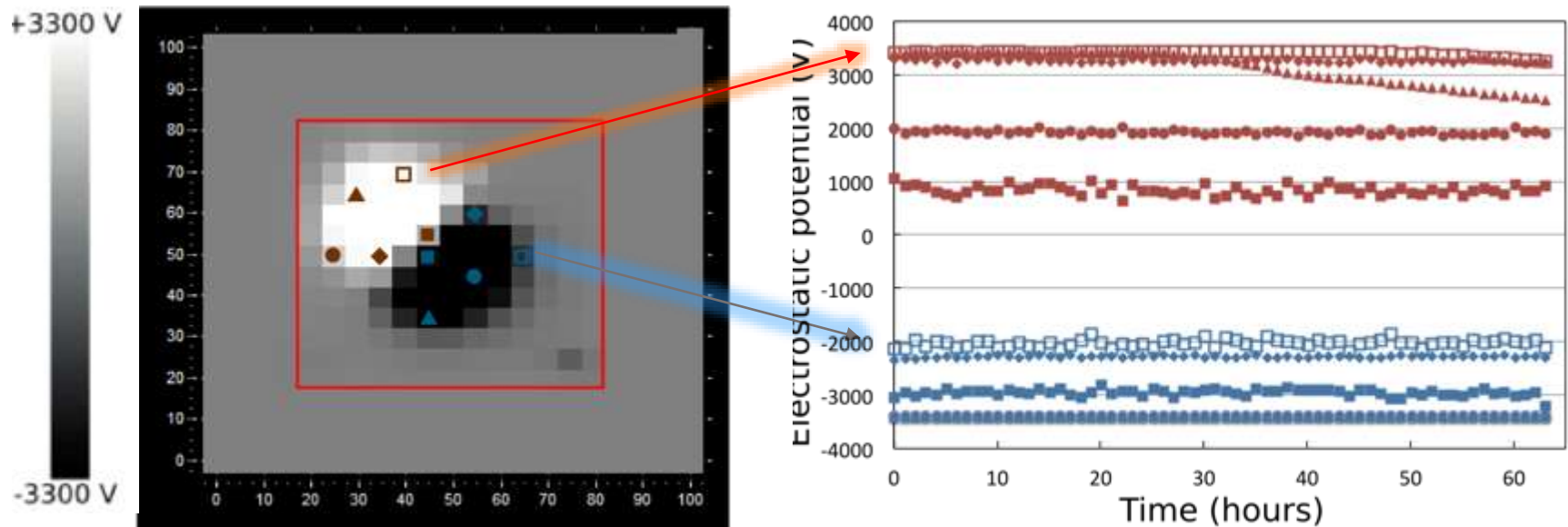


Stretching rubber exposes and hides sub-surface groups that adsorb water acquiring excess charge.

A capacitor is charged.

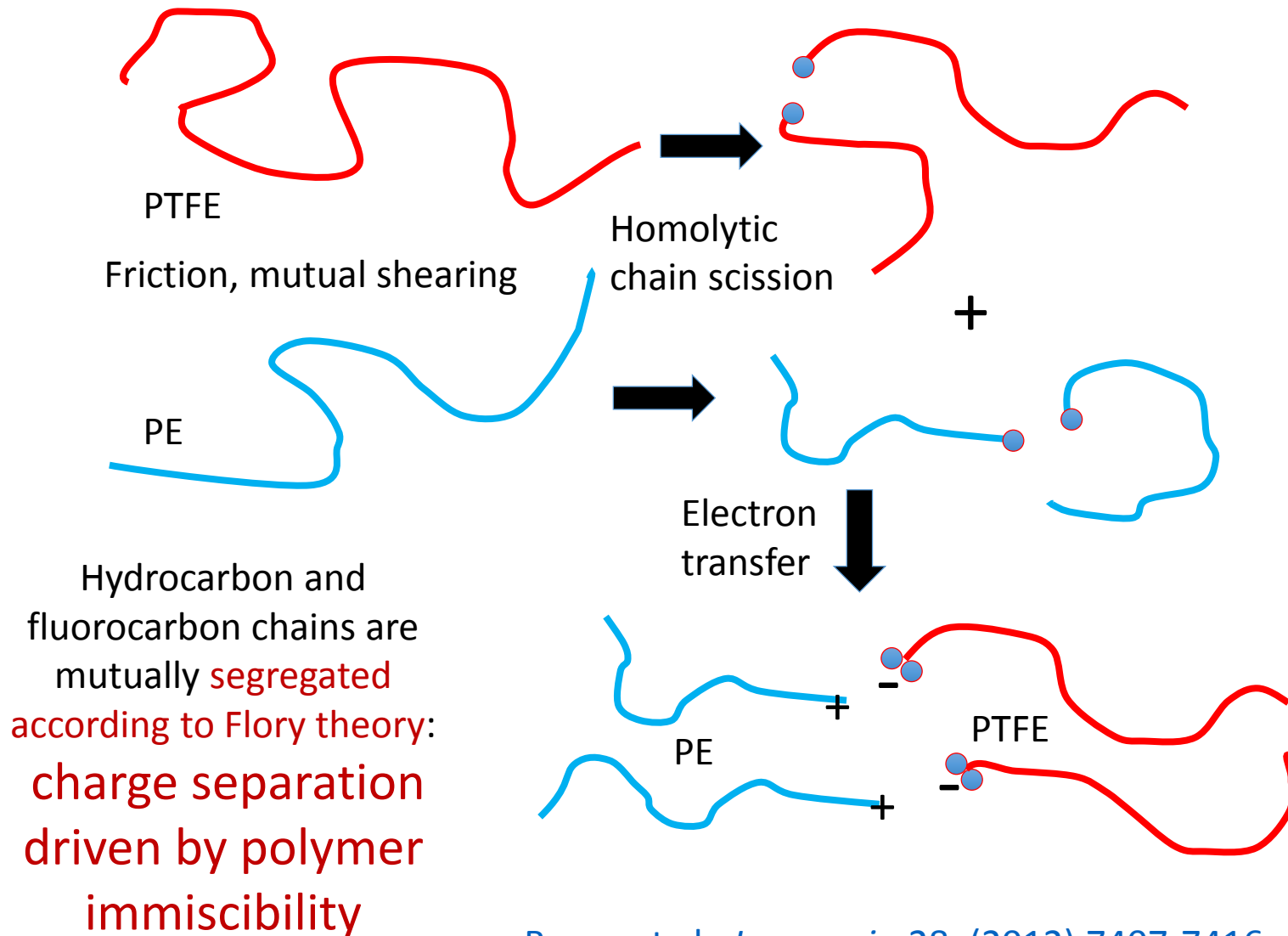
Burgo, Batista, Galembeck, ACS Omega (2018).

Charge produced by friction: PTFE rubbed with LDPE foam



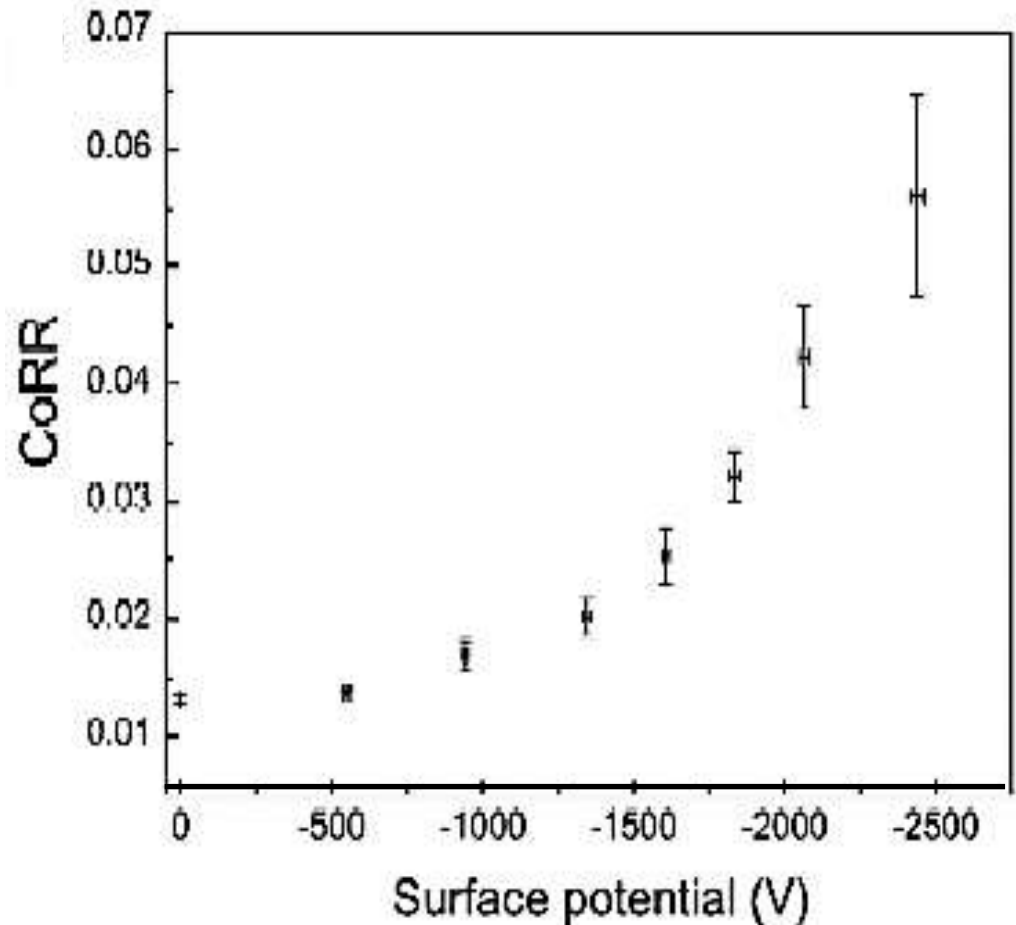
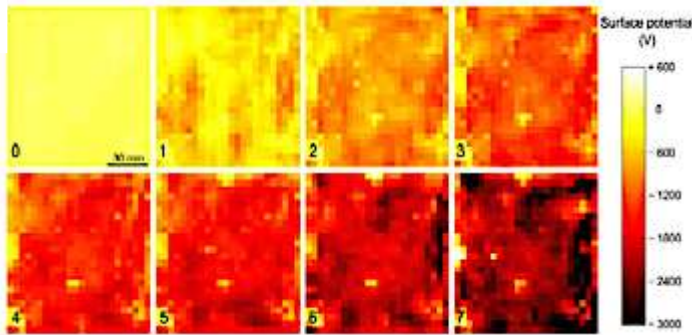
Formation of positive AND negative charges on the same surface.
Challenges the paradigm of unidirectional charge transfer in contacting solids.

How? mechanochemical mechanism for triboelectricity



Burgo et al., *Langmuir* 28, (2012) 7407-7416

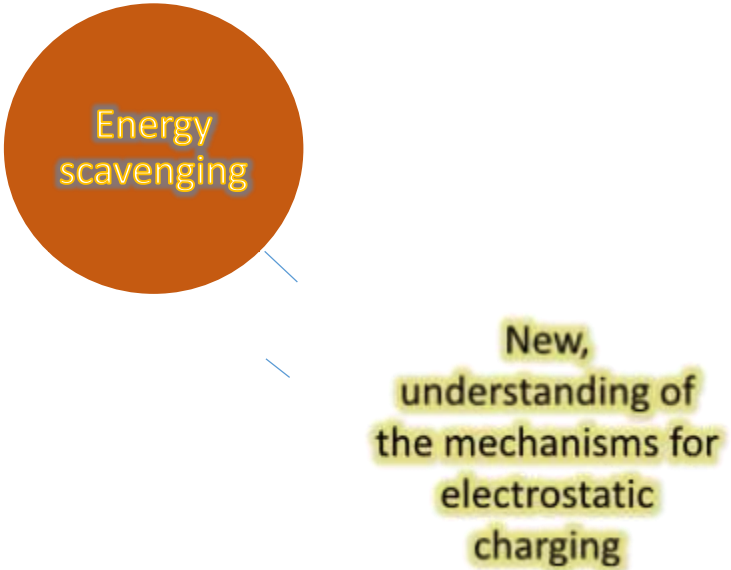
Feeding back: friction creates charge that **increases**
friction coefficients



T. A. L. Burgo, C. A. Silva, L. B. S.
Balestrin & F. Galembeck

Scientific Reports 3 (2013)
2384

Outlook



Energy
scavenging

New,
understanding of
the mechanisms for
electrostatic
charging

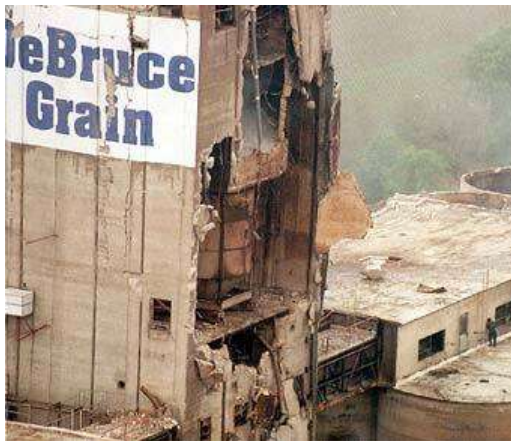
Electrostatics is Alive, Again

- We live in an **electrified environment**
- **Electricity is produced** within the Earth capacitor
 - **Electroneutrality** is the exception, anywhere
 - The charge **carriers**
 - **Chemical** electrostatics
- The atmosphere is a **charge reservoir**
 - Fires, explosions, safety
- Outlook: new science, new technology





Explosions triggered by electrostatic discharge

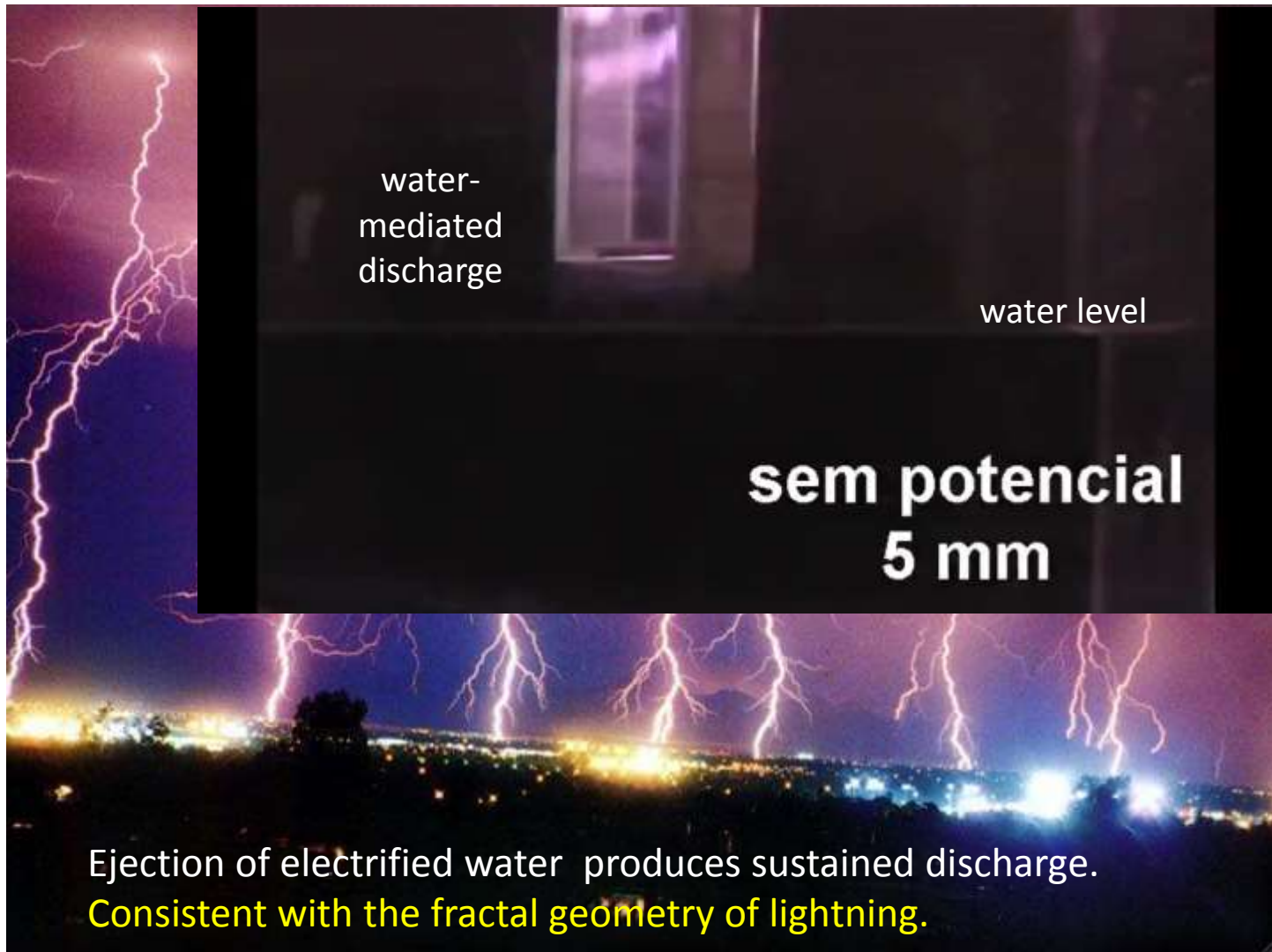


The first dust explosion that was outlined in a world literature occurred in 14 December 1785 in Italy.

Turin Science Academy noticed that it was an explosion of flour dust in the centre of Turin.

According to literature all building was destroyed.

Reported by Rafal Porowski from HQ of SFS. <http://www.ppoz.pl/wwwold/current.htm>



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A note on policy (II)

- Is money being spent where it should?
 - Little, poor diagnosis. Poor decisions on project implementation.
- Lack of assessment.
 - Results of the largest funding body in the country (FNDCT) do not even have a definite assessment methodology.
 - Poor planning
- Result: huge sums spent in facilities and equipment that do not provide the basis for the introduction of much-needed public policies.