

Non-Thermal Plasmas for Revolutionizing Goods Production

David B. Go

gogroup.nd.edu

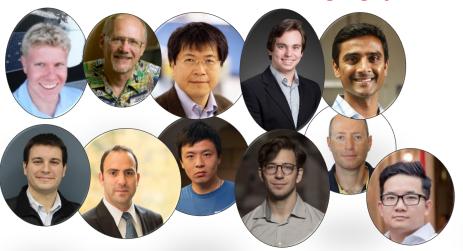
Aerospace and Mechanical Engineering Chemical and Biomolecular Engineering University of Notre Dame

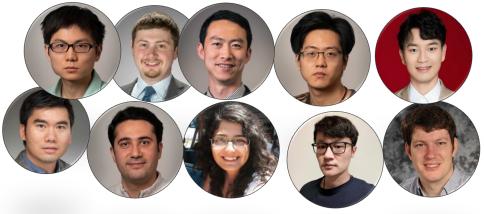
September 10, 2025

Plasma Electrochemistry

Prof. Paul Rumbach, Dr. David Bartels, Prof. Mohan Sankaran (Illinois), Prof. Chia Chang, Prof. Daniel Elg (Southern Indiana), Prof. Felipe Veloso (UC|Chile) Dr. Hernan Delgado, Dr. Jinyu Yang, Dr. Oles Dubrovski, Dr. Daniel Martin, Dr. Hoang Nguyen Plasma-Driven Material Processing

Prof. Yanliang Zhang, Prof. Alex Dowling, Prof. Tengfei Luo, Dr. Mike McMurtry (INL), Dr. Nazli Turan, Dr. Mortaza Saeidi-Javash, Dr. Jinyu Yang, Dr. Yipu Du, Dr. Ke Wang, Zhongyu Cheng





ARO W911NF-17-1-0119 ARO W911NF-23-1-0010 DOE Rickover Fellowship

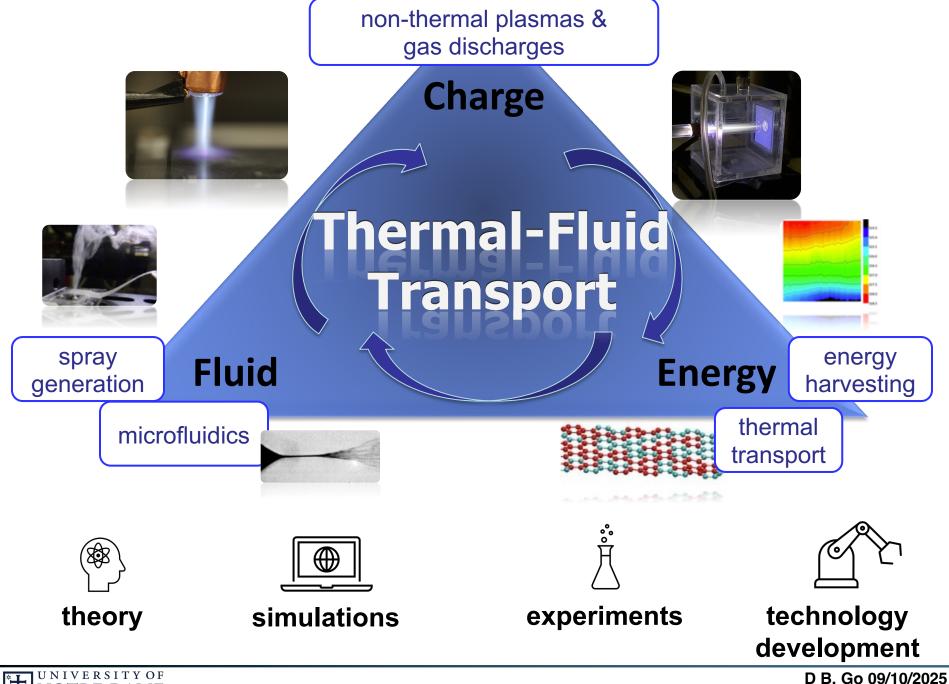
AFOSR FA9550-18-1-0157 DOE DE-EE0009103



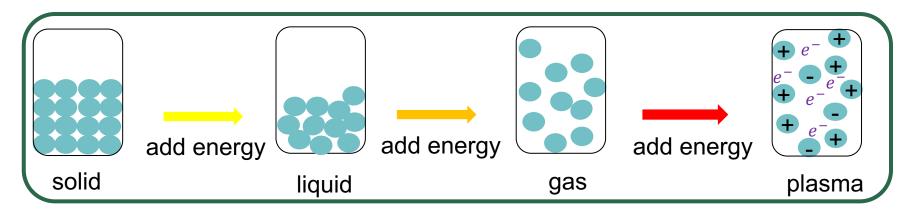








What is Plasma?





Thermal (equilibrium) Plasmas

Average T > 10,000 K $T_e \approx T_i \approx T_n$



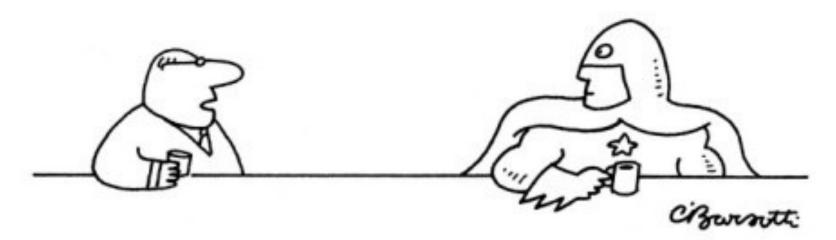


Non-thermal (non-equilibrium) Plasmas

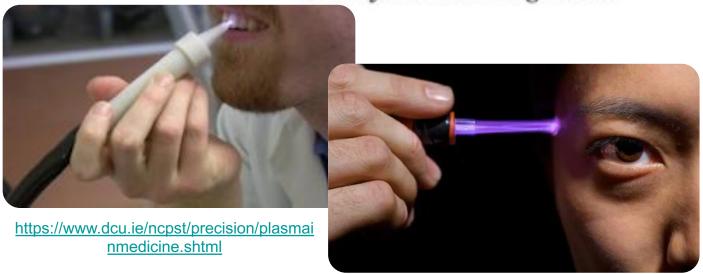
Average T ~ 300 K $T_e > T_i \approx T_n$



Non-Thermal Plasmas Revolutionizing Applications



"Some of us are unsung heroes."



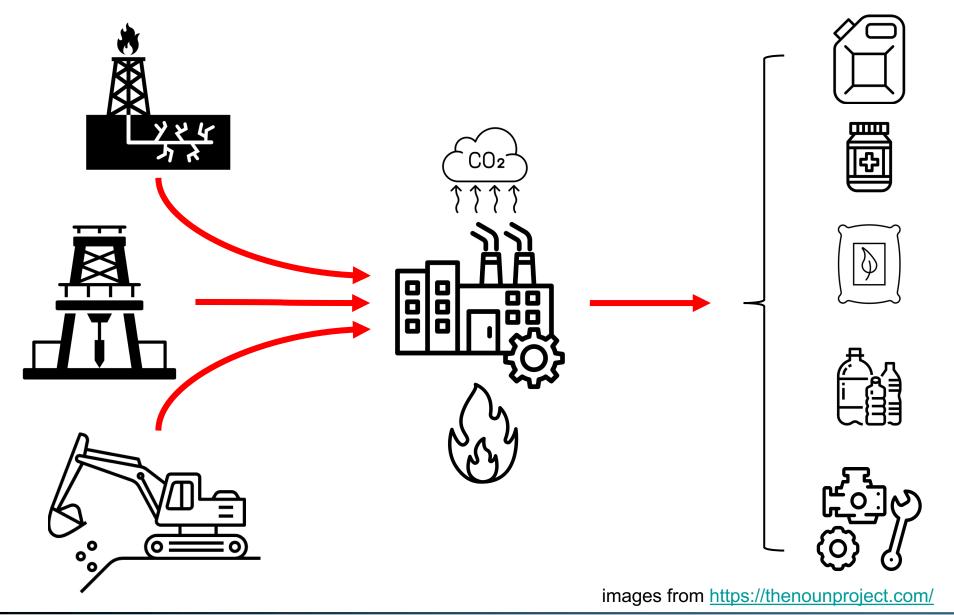
https://io9.gizmodo.com/now-you-can-replace-your-antibacterial-soap-with-a-plas-5899149



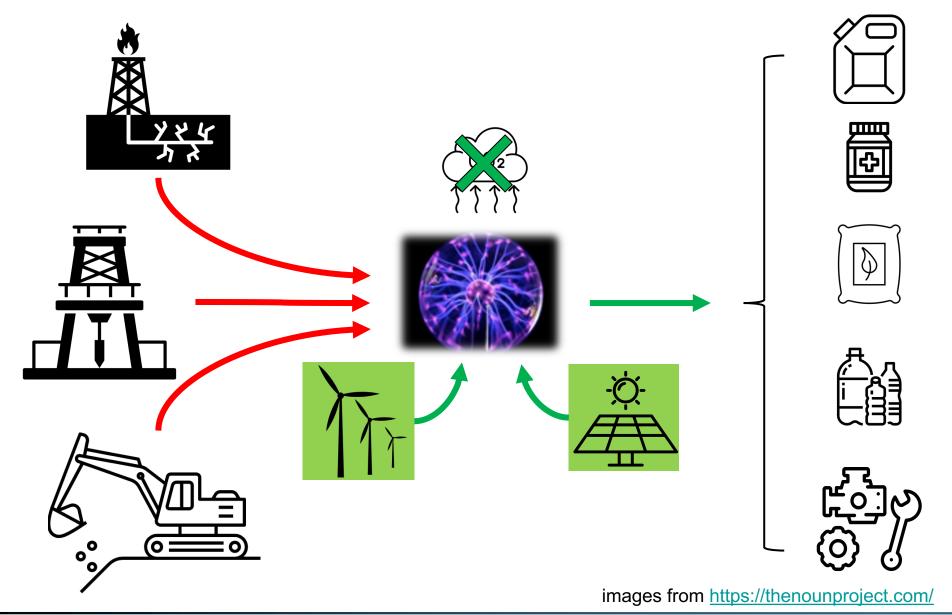
https://www.atbpotsdam.de/en/researchprograms/quality-and-safety-offood-and-feed/plasma.html



Plasmas for More Resilient Goods Production



Plasmas for More Resilient Goods Production



NSF WORKSHOP

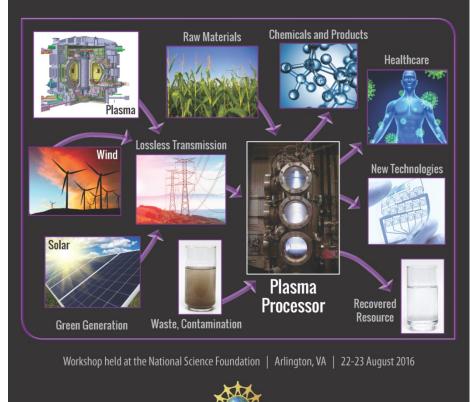
Science Challenges in Low **Temperature** Plasma Science and Engineering: **ENABLING A FUTURE BASED** ON ELECTRICITY THROUGH NON-EQUILIBRIUM PLASMA **CHEMISTRY**

Workshop report: arxiv.org/abs/1911.07076

> **Low Temperature Plasma** Science and Engineering



Enabling a Future Based on Electricity Through Non-Equilibrium Plasma Chemistry

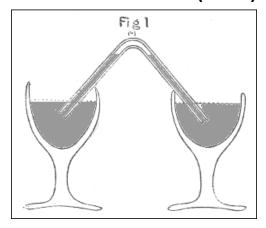


Courtesy: Mark Kushner, U. Michigan



Plasma-Liquid Systems As A Basis for Goods Production

Cavendish uses an arc to convert air to nitrous acid (1784)



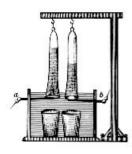
H. Cavendish, Experiments on Air, 1785.

Rayleigh and Ramsay use an arc to discover argon (1895)



escola.britannica.com.br

Schönbein uses an arc to discover ozone (1840)



courtesy J. Lopez, Seton Hall University

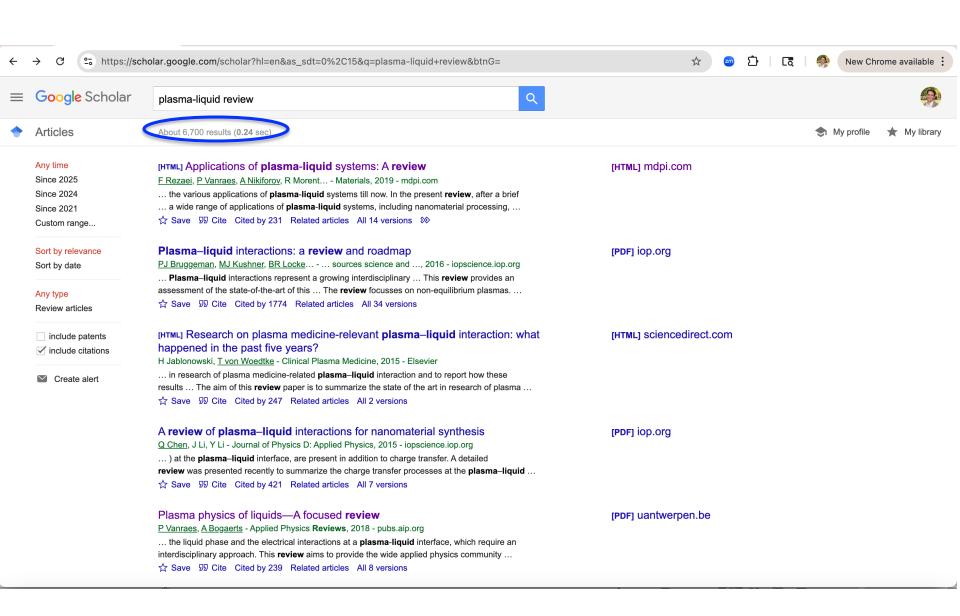
Birkeland-Eyde process to produce fertilizer patented (1903)





www.hydro.com/en/About-Hydro/Our-history/1900---1917/Explosive-winter-days-in-1903/





scitation.org/journal/jap

Plasma-liquid interactions

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Note: This paper is part of the Special Topic on Plasma-Liquid Interactions.

The Journal of Chemical Physics

REVIEW

pubs.aip.org/aip/jcp

Advances in plasma-driven solution electrochemistry

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Published Online: 19 February 2025

Peter J. Bruggeman, [16] ® Renee R. Frontiera, ® Uwe Kortshagen, ® Mark J. Kushner, ® Suljo Linic, ® © Ceorge C. Schatz, ® Himashi Andaraarachchi, ® Subhajyoti Chaudhuri, ® Han-Ting Chen, ® © Collin D. Clay, ® ® Tiago C. Dias, ® Scott Doyle, ® © Leighton O. Jones, ® Mackenzie Meyer, ® Chelsea M. Mueller, © Jae Hyun Nam, ® Astrid Raisanen, ® Christopher C. Rich, ® ©

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Journal of Applied Physics

PERSPECTIVE

scitation.org/iournal/iap

Cite as: J. Appl. Phys. **129**, 220901 (2021); doi: 10.1063/5.0044905 Submitted: 20 January 2021 · Accepted: 19 May 2021 · Published Online: 10 June 2021







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Note: This paper is part of the Special Topic on Plasma-Liquid Interactions.

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APPLIED PHYSICS REVIEWS 5, 031103 (2018)



APPLIED PHYSICS REVIEWS—FOCUSED REVIEW

Plasma physics of liquids—A focused review

Patrick Vanraes and Annemie Bogaerts

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(Received 22 December 2017; accepted 23 May 2018; published online 25 July 2018)

REVIEW ARTICLE

Charge transfer processes at the interface between plasmas and liquids

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(Received 14 March 2013; accepted 28 May 2013; published 19 June 2013)

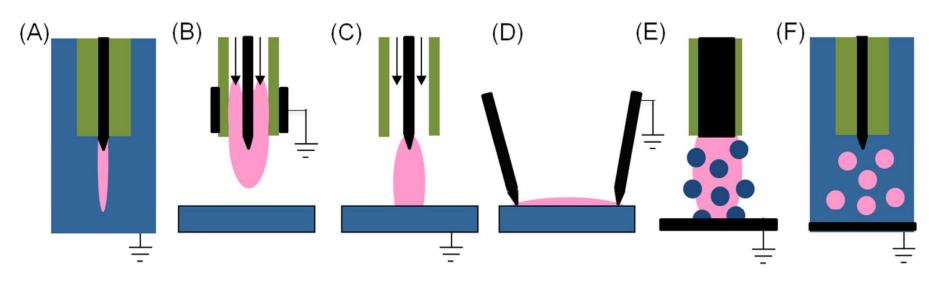


A Diverse Landscape of Interfaces

Discharge in liquid

Gas phase discharges

Multiphase discharges

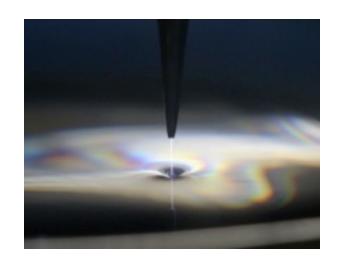


- A) Direct discharge in liquid
- B) Plasma jet
- C) Gas phase plasma with liquid electrode
- D) Surface discharge
- E) Gas discharge in aerosols
- F) Gas discharge in bubbles

Bruggeman et al. Plasma Sources Sci. Technol., 25, 053002 (2016).

Revolutionizing Goods Production: Two Stories

Plasma Electrochemistry





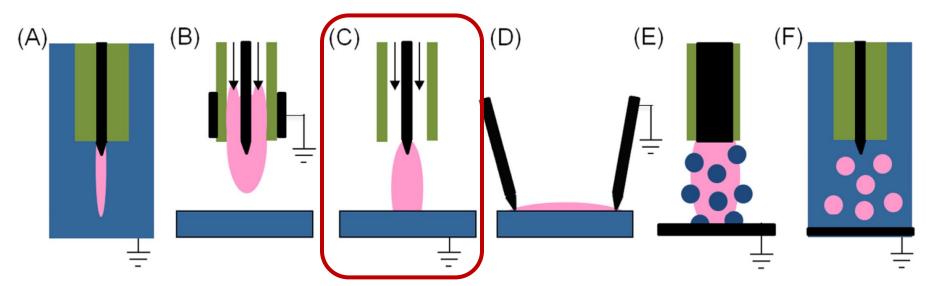
Plasma-Driven
Material Processing

A Diverse Landscape of Interfaces

Discharge in liquid

Gas phase discharges

Multiphase discharges

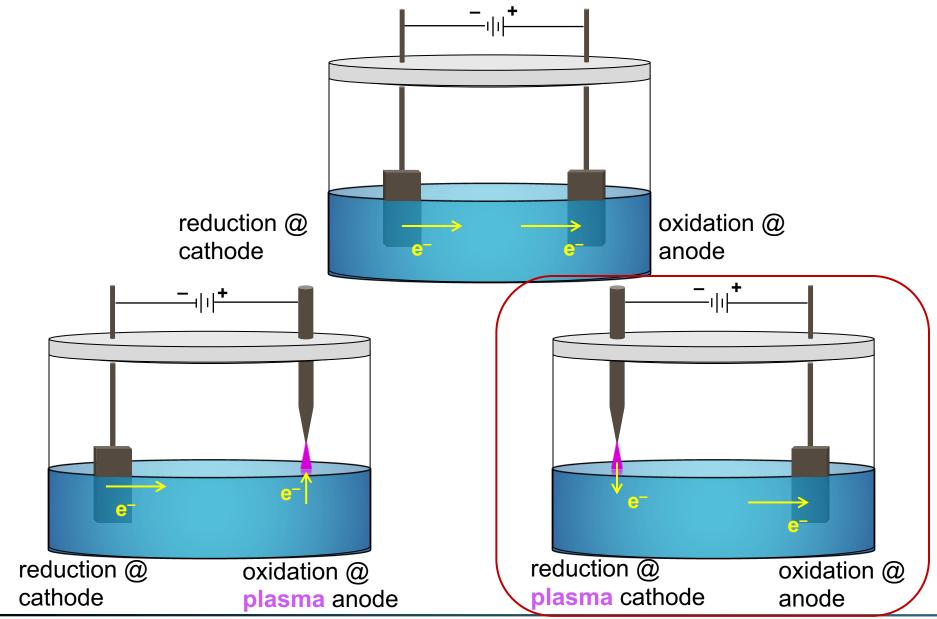


- A) Direct discharge in liquid
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Bruggeman et al. Plasma Sources Sci. Technol., 25, 053002 (2016).



Direct Plasma/Liquid Coupling



Plasma-Induced Water Electrolysis



Plasma-Injected Solvated Electrons



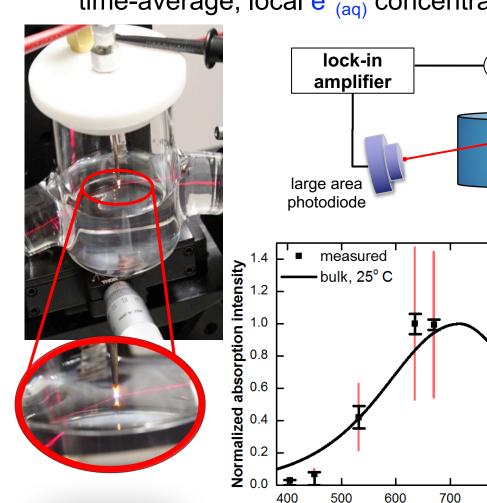
Measuring Plasma-Solvated Electrons

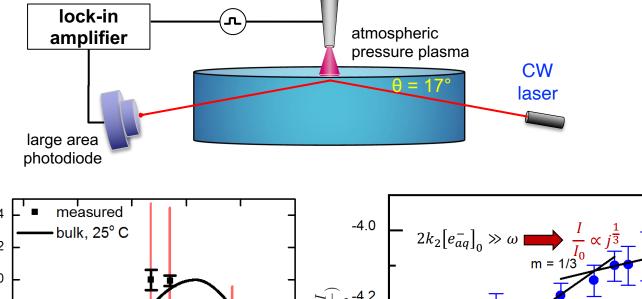


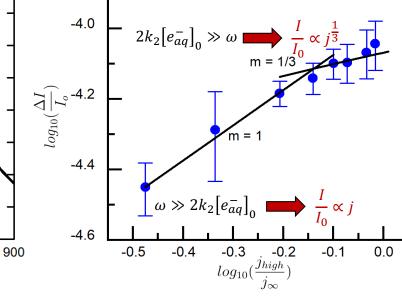




Total internal reflection absorption spectroscopy (TIRAS) measures the time-average, local e-(aq) concentration at the plasma-liquid interface







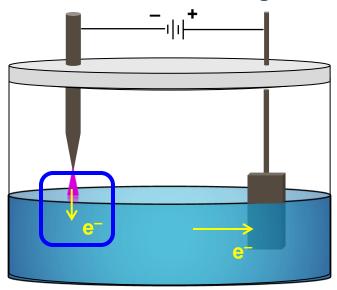


800

Laser wavelength (nm)

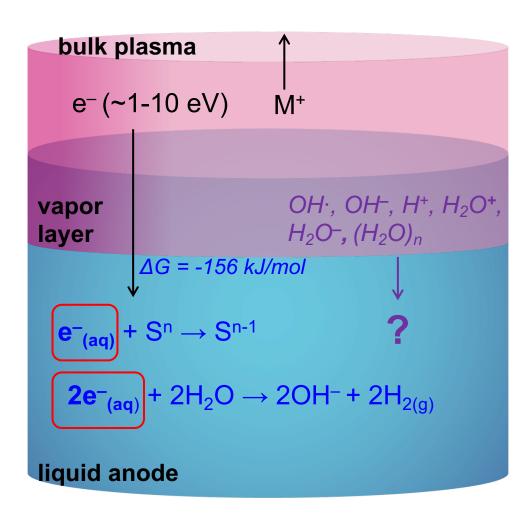
Solvated Electrons and the Plasma Cathode

plasma cathode configuration



Key Questions

- What chemistry can/do solvated electrons drive?
- What is the role of other plasmaproduced reactive species in the chemistry?
- How are the plasma and liquid coupled?



Spectrochimica Acta Part B: Atomic Spectroscopy 186 (2021) 106307



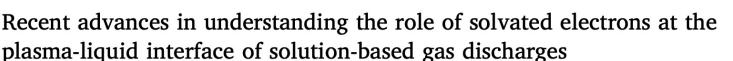
Contents lists available at ScienceDirect

Spectrochimica Acta Part B: Atomic Spectroscopy



journal homepage: www.elsevier.com/locate/sab

Review Article





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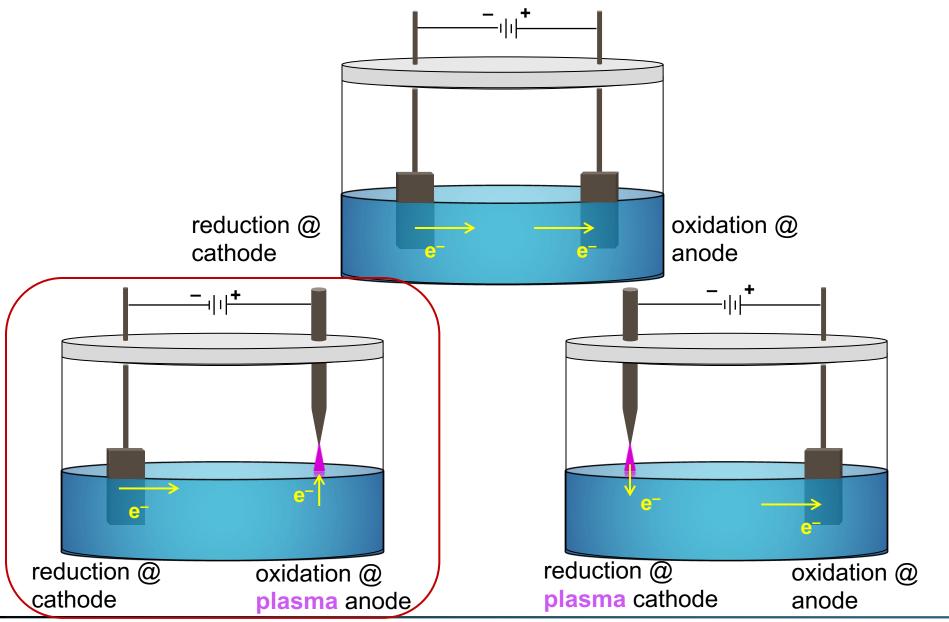
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^c Department of Aerospace and Mechanical Engineering, University of Notre Dame, Notre Dame, IN 46556, United States

d Department of Nuclear, Plasma, and Radiological Engineering, University of Illinois at Urbana-Champaign, Urbana, IL 61801, United States

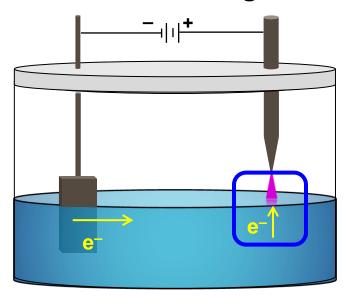
e Notre Dame Radiation Laboratory and Department of Chemistry and Biochemistry, University of Notre Dame, Notre Dame, IN 46556, United States

Direct Plasma/Liquid Coupling



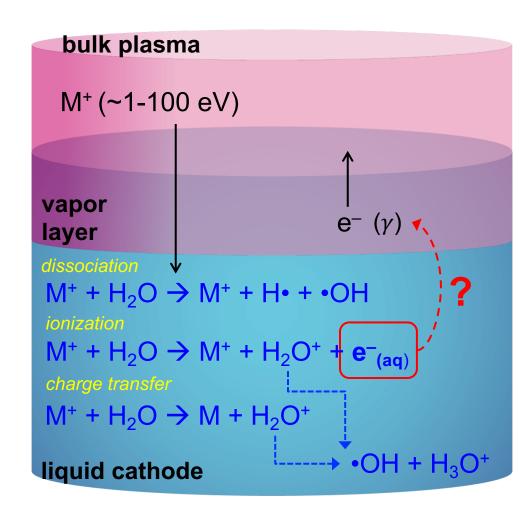
Solvated Electrons and the Plasma Anode

plasma anode configuration



Key Questions

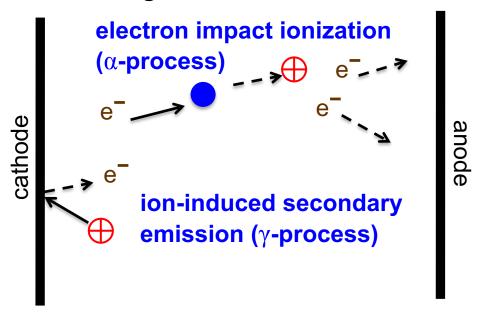
- What is the mechanism of secondary emission from a liquid and do solvated electrons play any role in this mechanism?
- What is the role of solvated electrons in this configuration?
- Can we detect solvated electrons?



Basics of Secondary Emission

In a plasma, **secondary emission** refers to the emission of electrons from the cathode from incoming positive ions

Charge Creation Processes



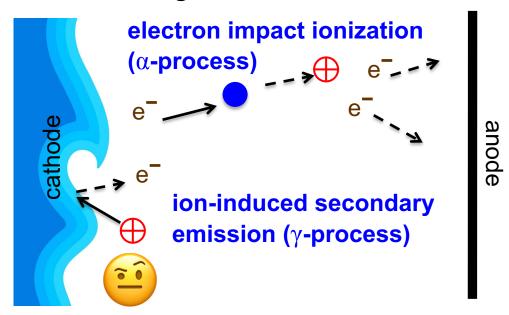
secondary emission coefficient

$$\gamma = rac{N_{e,emit}}{N_{p,incident}}$$

Basics of Secondary Emission

In a plasma, **secondary emission** refers to the emission of electrons from the cathode from incoming positive ions

Charge Creation Processes



secondary emission coefficient

$$\gamma = rac{N_{e,emit}}{N_{p,incident}}$$

Measuring Secondary Emission Coefficient



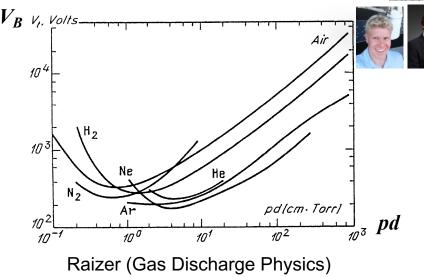


Paschen curve

$$V_{
m B} = rac{Bpd}{\ln(Apd) - \ln\Bigl[\ln\Bigl(1+rac{1}{\gamma_{
m se}}\Bigr)\Bigr]}$$

function of gas composition (A and B) and electrode material (γ)

$$\gamma = \frac{N_{e,emit}}{N_{p,incident}}$$



Modified expression for the Paschen curve (Spyrou et al. J. Phys. D. (1994))

$$V_B' \, \exp\left(-B \, \frac{R}{2d} \, \frac{pd}{V_B'}\right) \left[1 - \exp\left(-B \, \frac{pd}{V_B'}\right)\right] = \frac{B}{A} \mathrm{ln} \left(1 + \frac{1}{\gamma}\right)$$

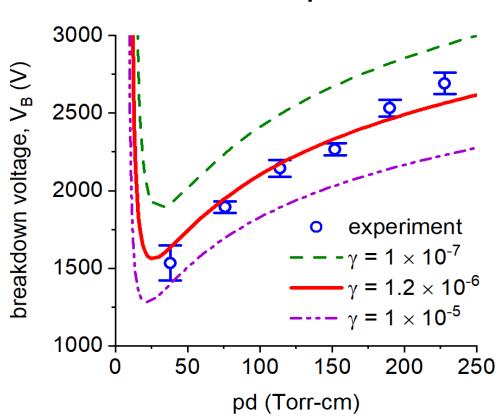
where,

$$V_B' = \frac{V_B}{\ln\left(1 + 2d/R\right)}$$

Estimate γ by measuring V_B at different pd values

Small Fraction of Bombarding Ions Result in an Electron Being Emitted

Breakdown voltage as a function of pd

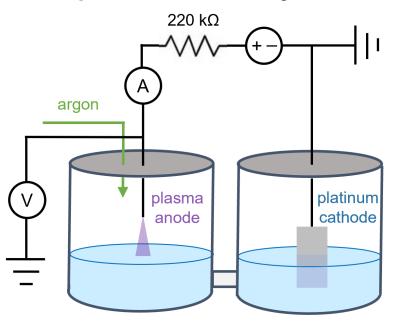


Best fit $\rightarrow \gamma = 1 \times 10^{-6}$

- maximum γ of ~ 10⁻⁵
- secondary emission mechanism is very inefficient

Plasma Voltage Indicator of the Secondary Emission

split cell reaction system



system characteristicss

- argon headspace
- 10 mA current
- 1 mm capillary-liquid separation

If a critical variable for secondary emission is removed, the gas might not transition into a plasma

Chemical Scavengers Remove Potential Contributors to Secondary Emission

Nitrite (NO₂-)

$$e_{aq}^{-} + NO_{2}^{-} \rightarrow NO_{2}^{2-}$$

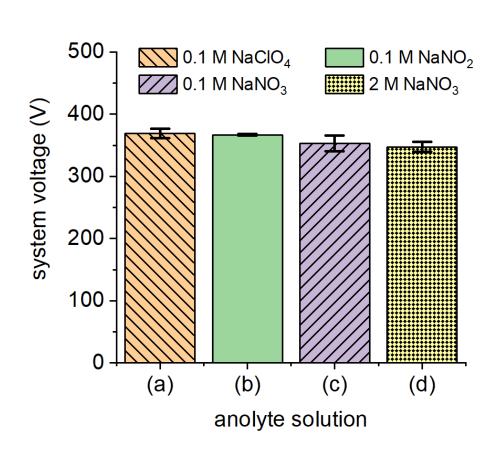
$$H \cdot + NO_{2}^{-} \rightarrow OH^{-} + NO$$

$$\cdot OH + NO_{2}^{-} \rightarrow OH^{-} + \cdot NO_{2}$$

Nitrate (NO₃-)

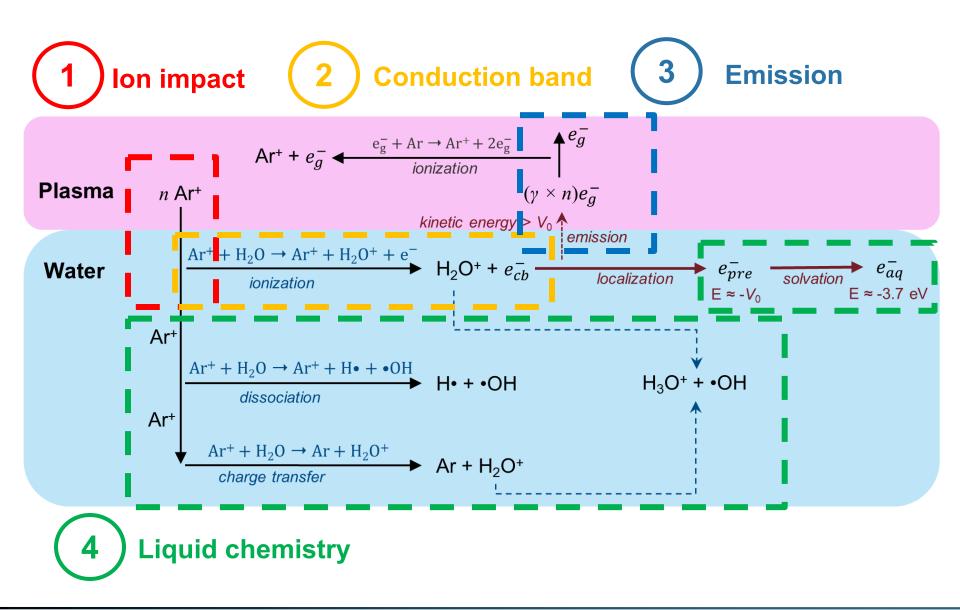
$$e^-_{aq} + NO_3^- \rightarrow NO_3^{\; 2-}$$

$$e_{pre}^{} + NO_3^{-} \rightarrow NO_3^{2-}$$

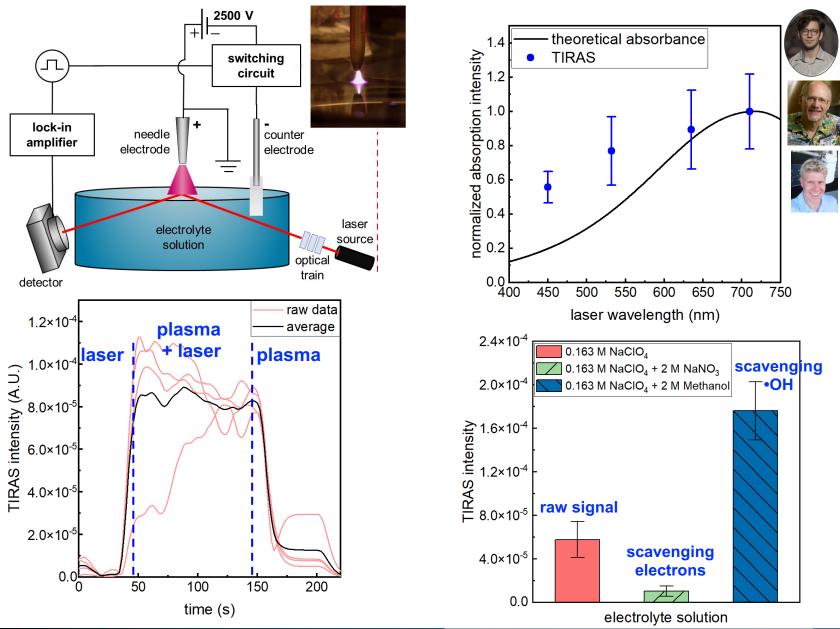


- plasma-liquid chemistry has no effect on system voltage
- emitted electron is never solvated or pre-solvated

Possible Process for Secondary Emission



Do We Even Know There Are Solvated Electrons?





How Many Ions Produce a Solvated Electron?

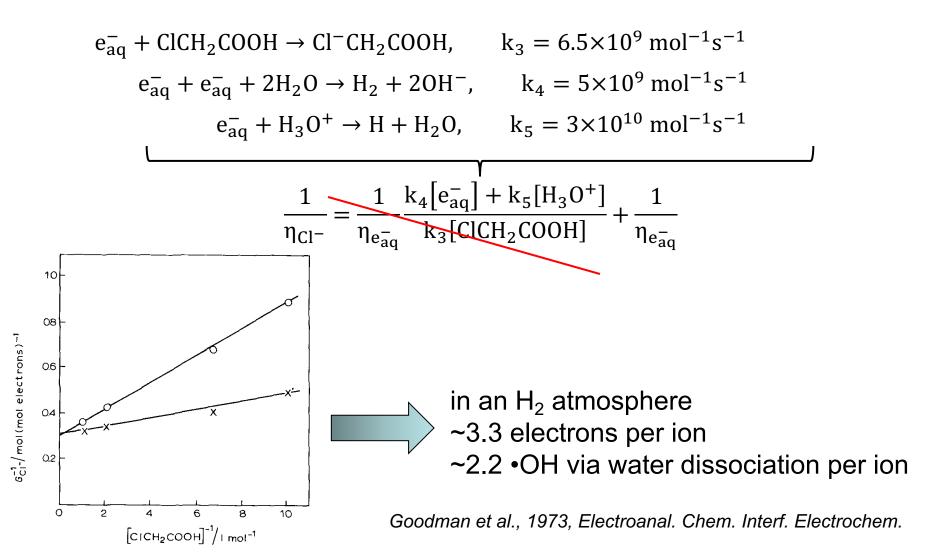
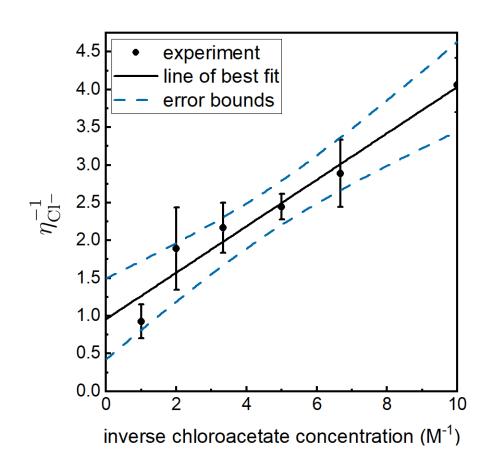


Fig. 1. Plot of G_{Cl}^{-1} versus [ClCH₂COOH]⁻¹, in (\bigcirc), 0.5 M H₂SO₄; (\times), H₂O.

Faradaic Efficiency of the Plasma Anode





in an Ar atmosphere ~1 electron per ion ~ 2 •OH per ion

$$\eta^{-1} = (0.308 \pm 0.098)x + (0.958 \pm 0.543)$$

$$\eta = 1.04 \pm 0.59 \left[\frac{e^{-1}}{\text{incident ion}} \right]$$

Plasma Electrochemistry for Uranium Extraction

Broad need to recover uranium from water

- in situ leach (ISL) mining (56% of world's uranium mining)
- environmental decontamination (mining & weapons programs)
- extraction from seawater



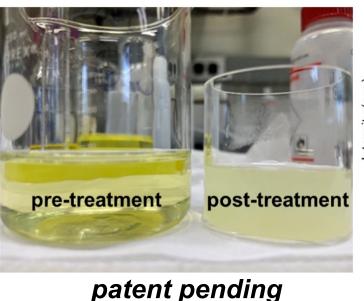


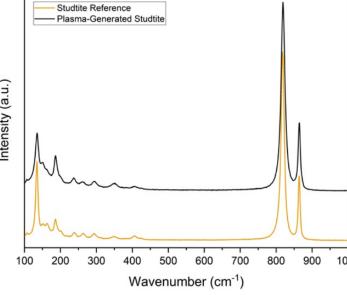
Typically achieved by reacting uranyl with hydrogen peroxide

$$UO_2^{2+} + 4H_2O + H_2O_2 \rightarrow [(UO_2)(O_2)(H_2O_2)_2](H_2O_2)_2 + H_2$$

uranyl (sulfate or carbonate) uranyl peroxide (studtite)

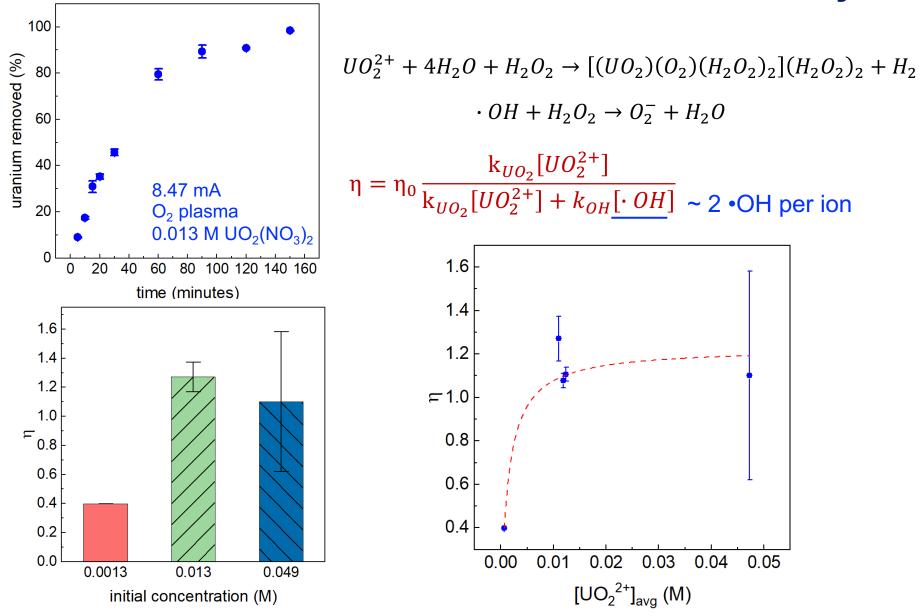








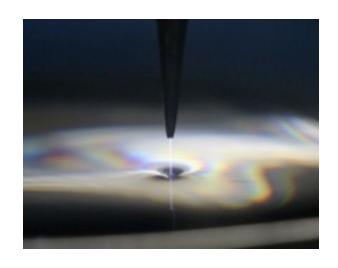
Extraction Can Achieve 100% Faradaic Efficiency





Revolutionizing Goods Production: Two Stories

Plasma Electrochemistry





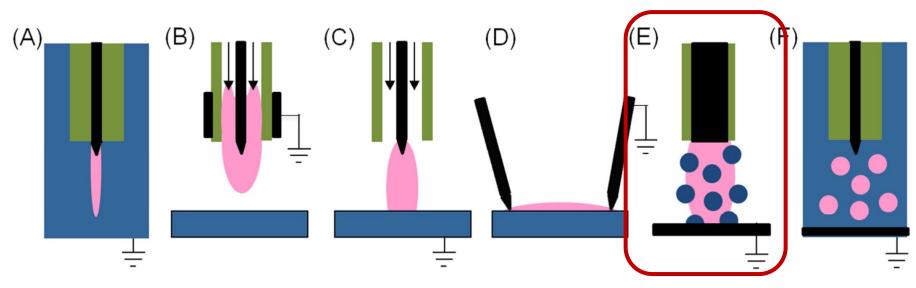
Plasma-Driven
Material Processing

A Diverse Landscape of Interfaces

Discharge in liquid

Gas phase discharges

Multiphase discharges



- A) Direct discharge in liquid
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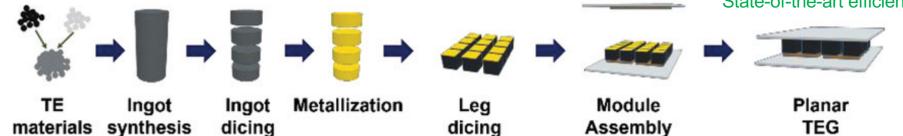
Bruggeman et al. Plasma Sources Sci. Technol., 25, 053002 (2016).



Advanced Manufacturing of Thermoelectrics

Bulk material-based manufacturing of thermoelectric modules

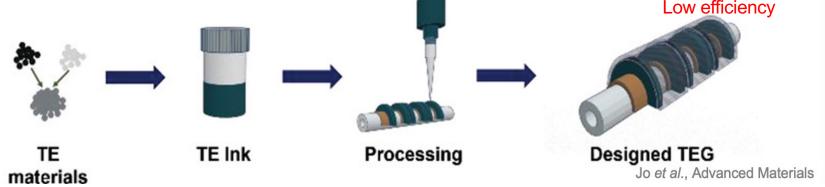
Time consuming
Uses more material
Large material waste
Assembling challenges
State-of-the-art efficiency



semanticscholar.org

Ink-based additive manufacturing for thermoelectric modules

Needs less material Little material waste No assembling needed 10× larger power density 90% cost reduction Low efficiency



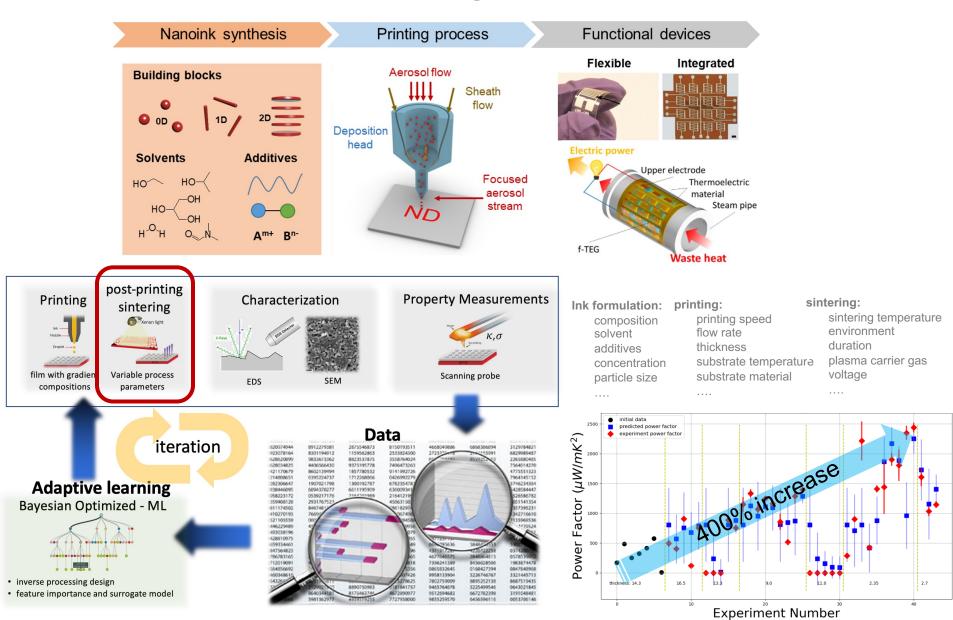




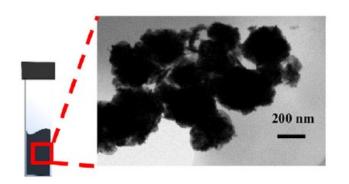




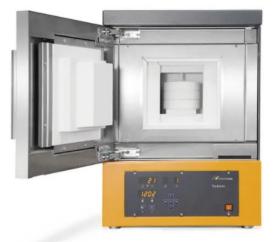
Data-Driven Manufacturing Optimization



Can Non-Thermal Plasmas Tackle Sintering?



Danaei, R. et al. Advanced Engineering Materials 21, 1800800 (2019)



https://www.medicalexpo.com/

oven sintering

nanoink preparation

print thin film

high temperature-activation for several hours

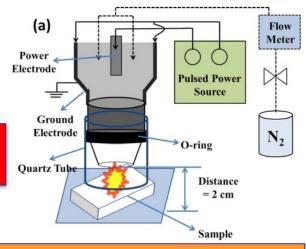
- > 400 °C
- 45 min 3 h

UNIVERSITY OF Kamyshny et al., Small, **10**, 3515–3535, 2014

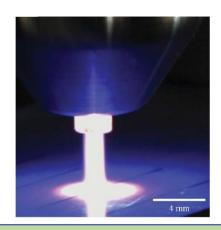
Can Non-Thermal Plasmas Tackle Sintering?



N₂ plasma sintering of AlN/CNT Substrate temperature = 700 °C



N₂ plasma sintering of TiO₂ Substrate temperature = 500 °C



Ar plasma sintering of Ag Substrate temperature < 70 °C

700 °C

500 °C

400 °C

50 °C

Substrate Temperature

Chiu, Y.-F. et al. Applied Surface Science 377, 75–80 (2016)

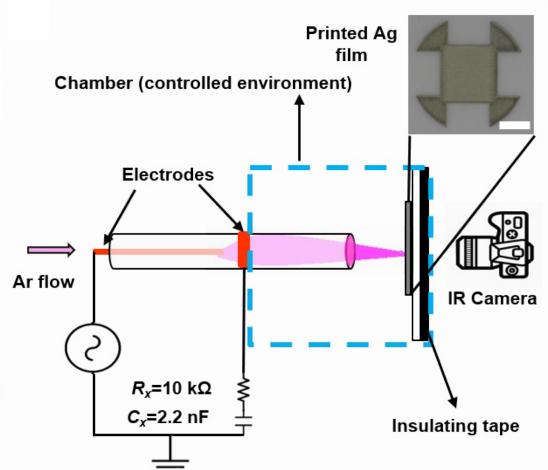
Wang, C. et al. ECS Journal of Solid State Science and Technology 4, P3020–P3025 (2015) Wünscher, S. et al. Journal of Materials Chemistry 22, 24569 (2012)

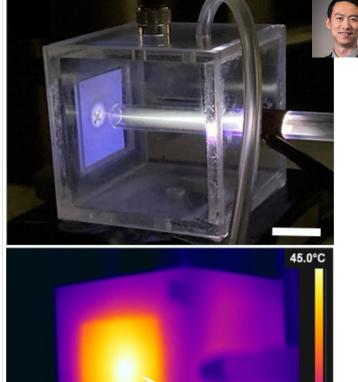


Plasma Jets for Low-Temperature Sintering









Active

plasma area

Substrates:

- Glass
- PEL paper

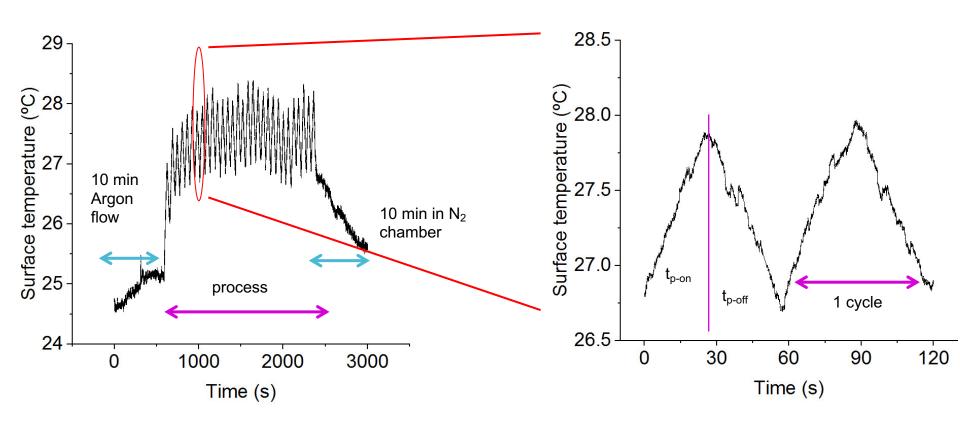
Gas composition:

- Flowing: 1500 sccm Ar
- Chamber: 10 sccm N₂

Thin films: 1-layer printed with Clariant's PRELECT® TPS Nano Silver Conductive Ink (30-50 nm particles)

22.9°C

Modulation for Near Room Temperature Conditions



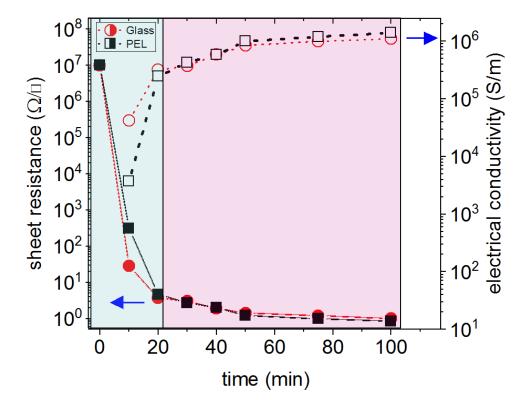
- > Trade-off: The cooling down time during the plasma-off portion of the cycle does not affect sintering or the conductivity considerably
- > Advantage: Sintering on temperature-sensitive components

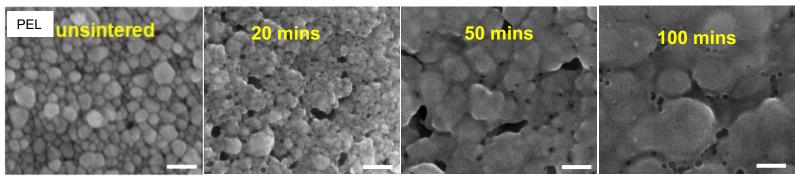


Plasma Jet Sintering Achieved

First Phase (first 20 min): removal of surfactants in the ink by energetic plasma ions and electrons that dissociate chemical stabilizers

Second Phase (20-100 min): densification driven by diffusion processes



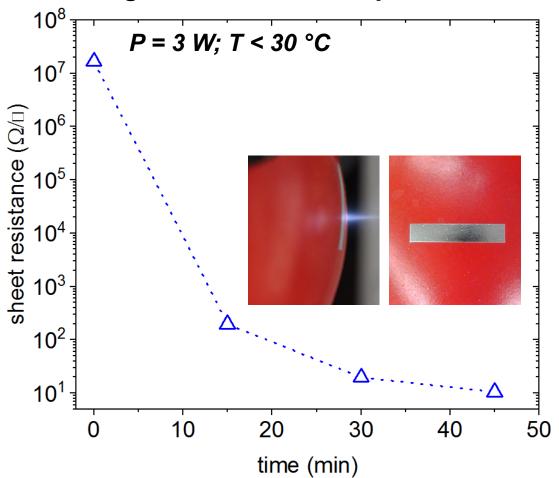


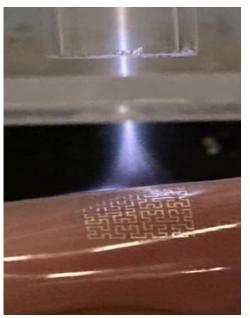
Scale bar:50 nm

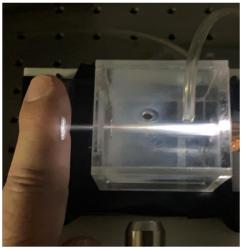


Plasma Jet Sintering on 'Delicate' Substrates

sintering on the flesh of a ripe tomato



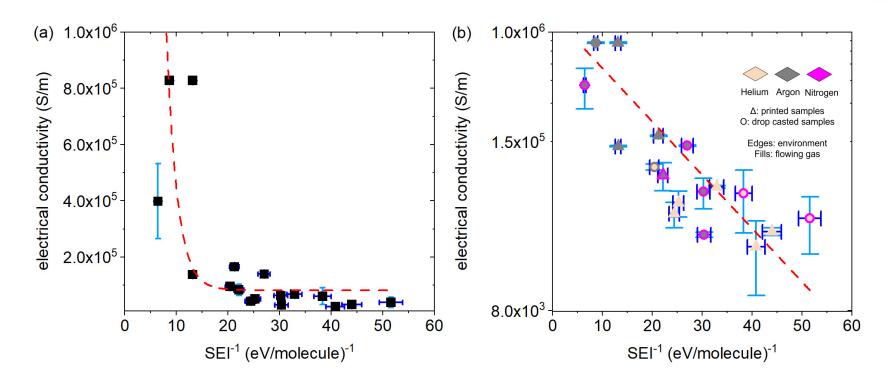




Evidence for Arrhenius-Like Relationship



the electrical conductivity of the film after 2 min of plasma exposure for 17 different SEI conditions



- The most effective conditions occur at high SEI
- \triangleright Linear fit of the data shows that the electrical conductivity (after 2 min of sintering) follows an Arrhenius-like $\exp(-\frac{E_c}{SEI})$ relationship



Plasma-Integrated Aerosol Jet Printer?



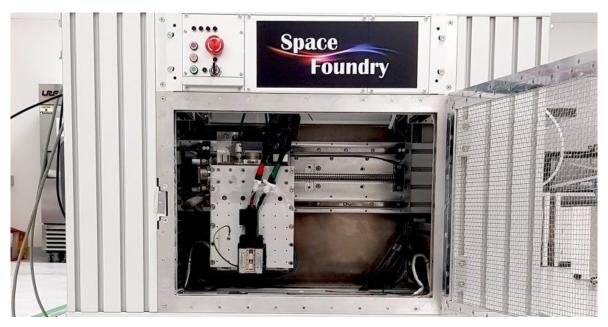
Research Article

www.acsami.org

Plasma Jet Printing of Electronic Materials on Flexible and Nonconformal Objects

Ram P. Gandhiraman,* Vivek Jayan, Jin-Woo Han, Bin Chen, Jessica E. Koehne, and M. Meyyappan

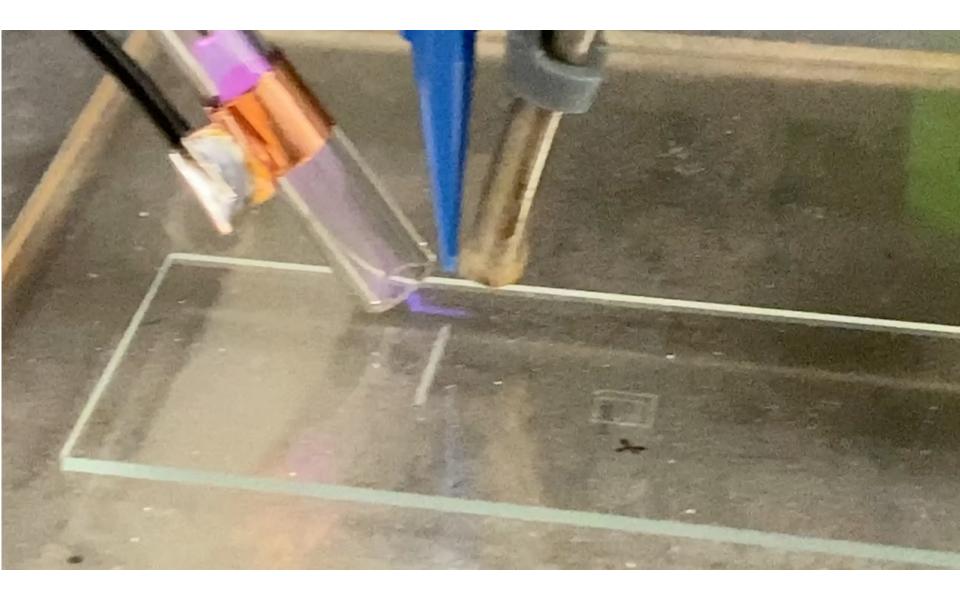
NASA Ames Research Center, Moffett Field, California 94035, United States



www.spacefoundry.us



First Generation: Parallel Jets

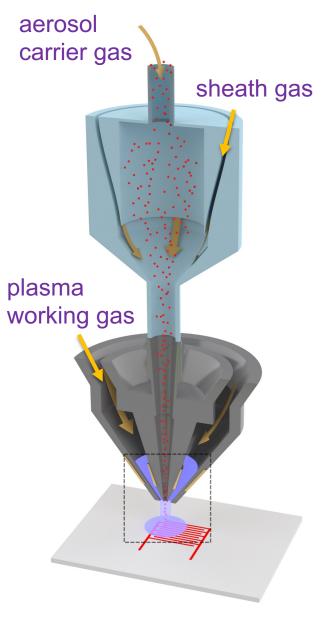


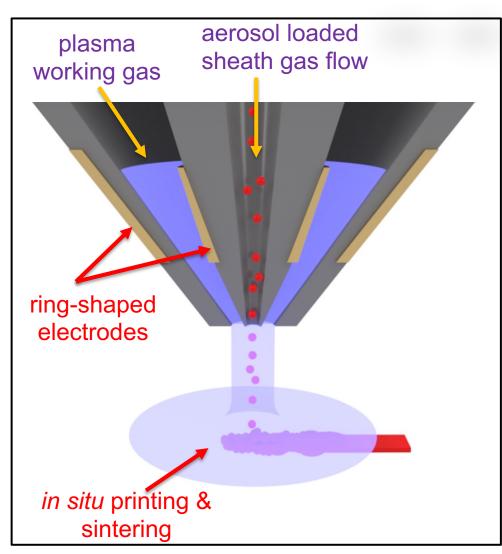
Second Generation: Co-Axial Jets







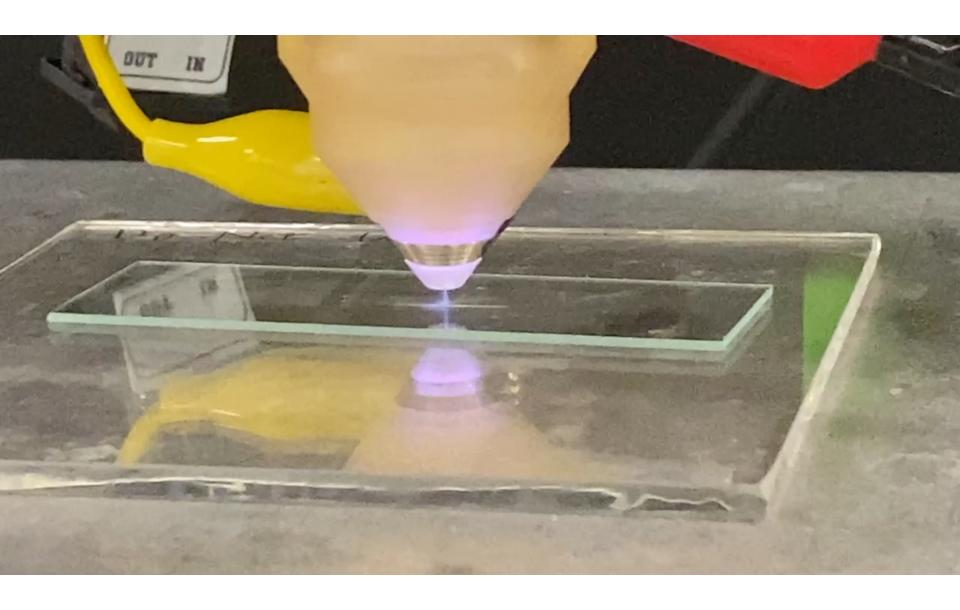




patent pending

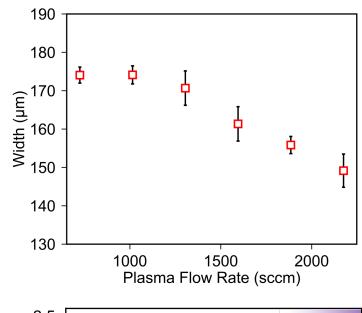


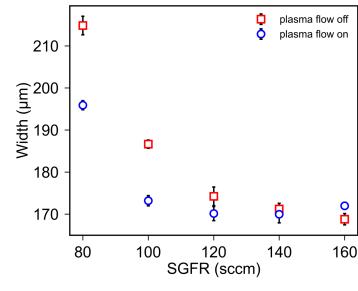
Second Generation: Co-Axial Jets



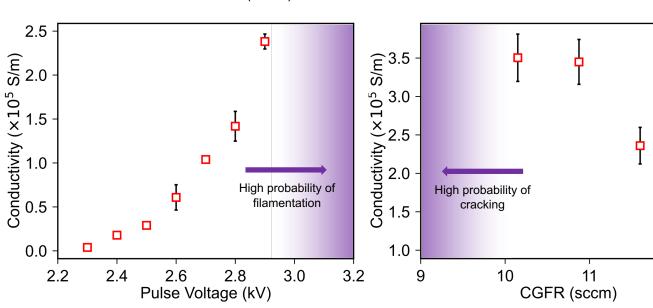
Parameter Variation: Resolution & Performance

 Both plasma gas and sheath gas affect print resolution





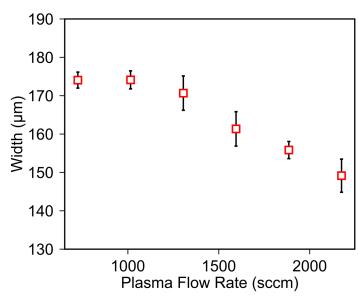
Challenge to inhibit cracking of the film as it leads to low conductivity and plasma-driven damage

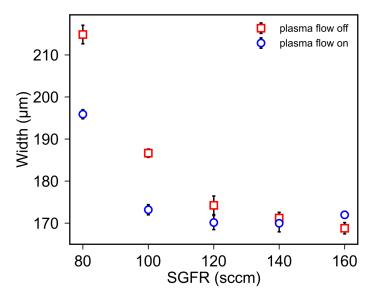


12

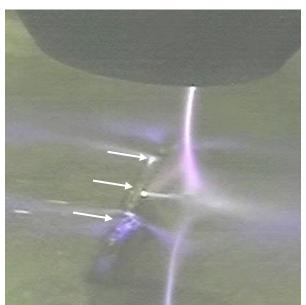
Parameter Variation: Resolution & Performance

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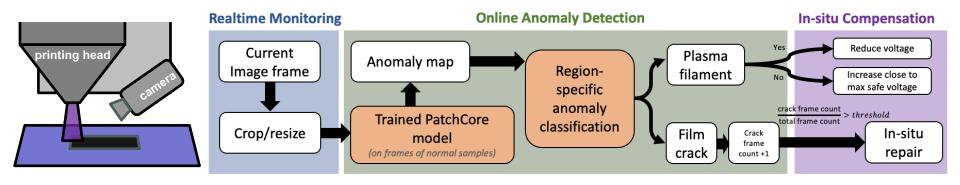


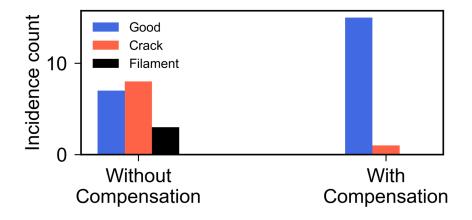


Challenge to inhibit cracking of the film as it leads to low conductivity and plasma-driven damage

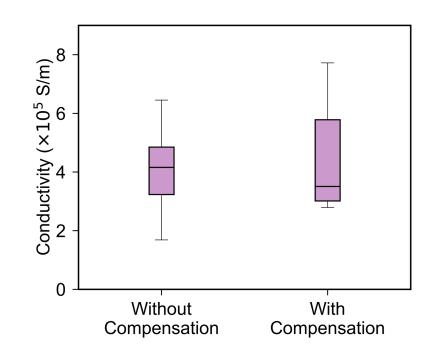


ML-Based Real Time Control



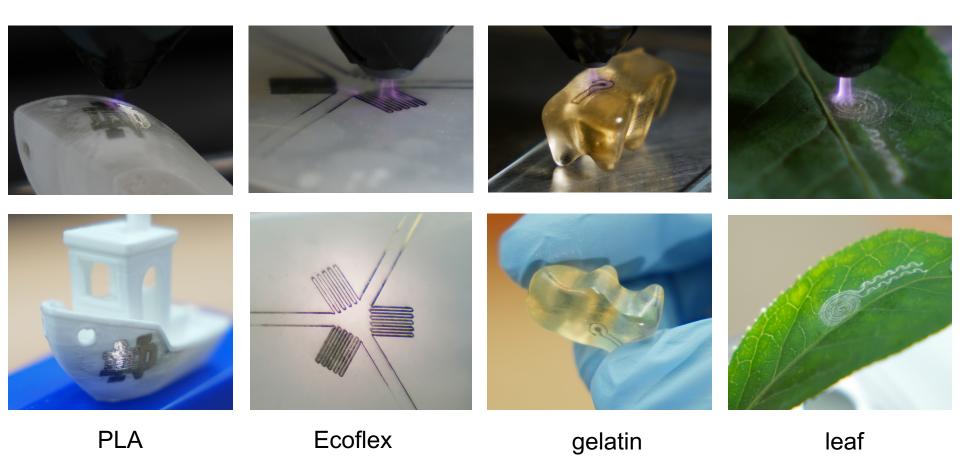


ML-enabled monitoring allows for higher yield with no impact on performance





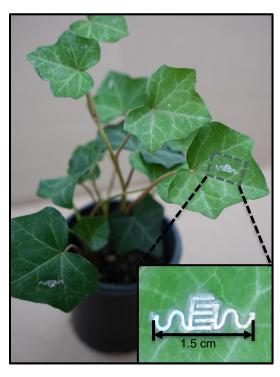
Co-Jet Printing: Rapid, Conformal, Versatile



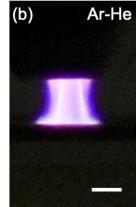


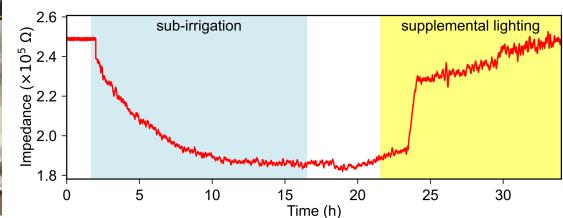
The Potential for Functional Devices





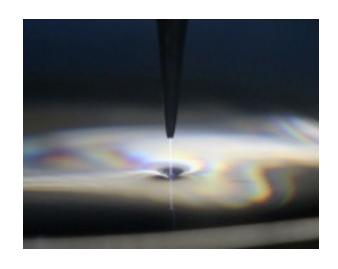






Revolutionizing Goods Production: Two Stories

Plasma Electrochemistry



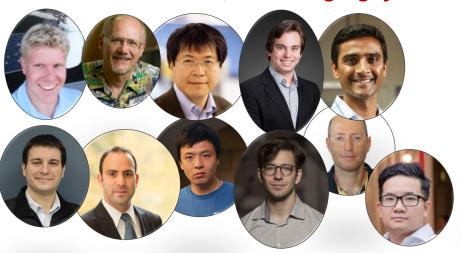


Plasma-Driven
Material Processing

Plasma Electrochemistry

Prof. Paul Rumbach, Dr. David Bartels, Prof. Mohan Sankaran (Illinois), Prof. Chia Chang, Prof. Daniel Elg (Southern Indiana), Prof. Felipe Veloso (UC|Chile) Dr. Hernan Delgado, Dr. Jinyu Yang, Dr. Oles Dubrovski, Dr. Daniel Martin, Dr. Hoang Nguyen Plasma-Driven Material Processing

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AFOSR FA9550-18-1-0157 DOE DE-EE0009103







