Multiscale Modeling of Plasma Water Interfaces

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Outline

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    2. Species in the gas and liquid phase
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Motivation: Plasma-Water Interactions

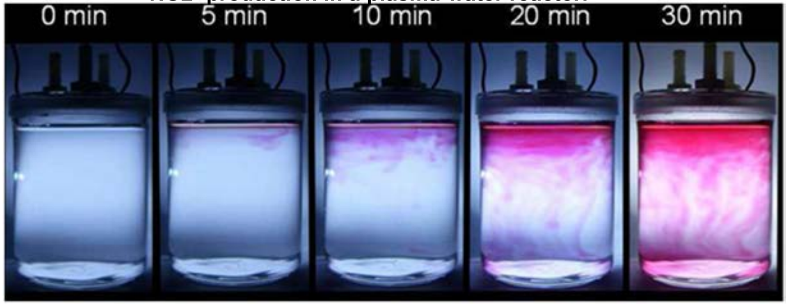
Applications

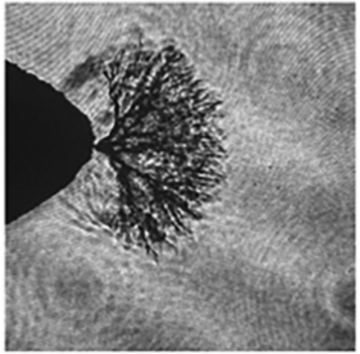
- **Generation of reactive oxygen and nitrogen species**
  - Antimicrobial properties
  - Ammonia production
  - Wound disinfection and healing
  - And more
- **Other, domain specific**
  - Plasma medicine
  - Synthesize graphene particles and nanosheets
  - Toxic metal detection
  - And much more

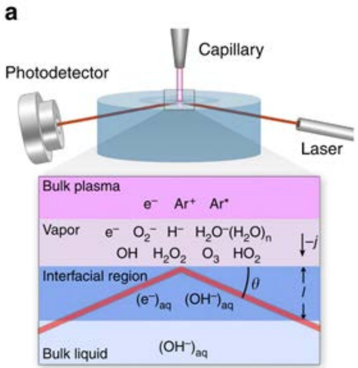
Advantages

- **Cheap and abundant materials**
- **“Cold” plasma - useful for thermally sensitive surfaces**
  - heat-sensitive equipment
  - bodily wounds

NO2- production in a plasma-water reactor.







Top: NO2- production in a plasma-water reactor. [1]
Bottom-right: Schematic of solvated electron measurement experiment. [2]
Bottom-left: Streamers propagating in liquid water. [3]

[1] Oehmigen K. IEEE Trans. Plasma Sci. 39 2646 (2011)
[2] Rumbach, P. *et al.* Nature Communications 6 7248 (2015)
[3] Foster, J., Physics of Plasmas 24 055501 (2017)

Methods

- **Plasma-in-liquid**
  - Directly ionize water phase with high voltages
  - Requires high voltages, but good source of OH production
- **Bubble plasmas**
  - Gas composition of bubbles may be tailored to adjust chemistry
- **Plasma-liquid interface**
  - Plasma generated in gas phase
  - Transport of reactive species depends on diffusion through water interface
  - Electrons drive RONS production by entering water phase and solvating

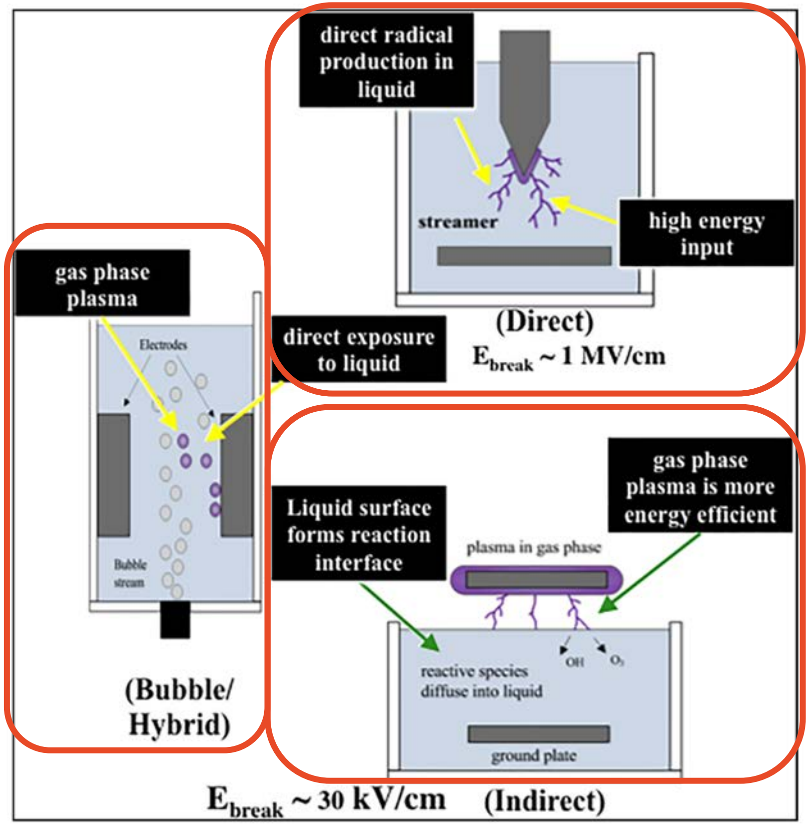
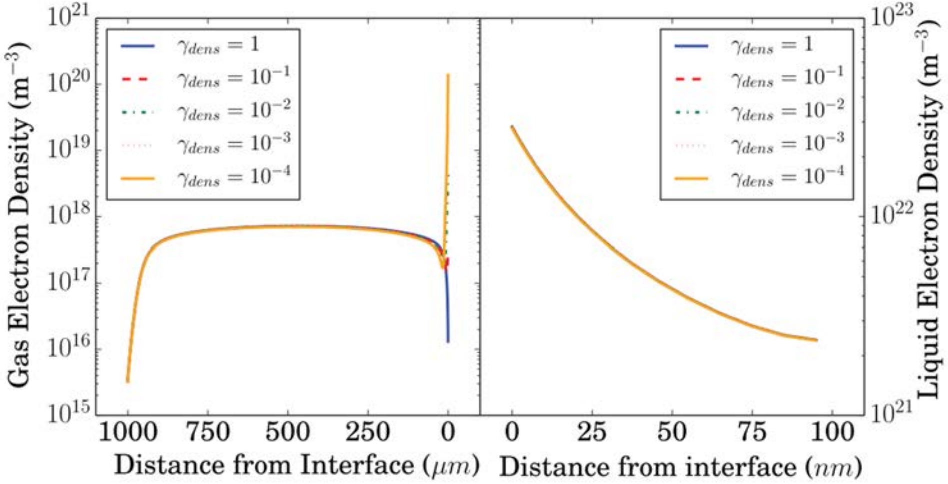


Figure adapted from [3].

[3] Foster, J. Plasma-based water purification: Challenges and prospects for the future." Physics of Plasmas 24 055501 (2017)

Introduction to Zapdos-Crane

- **Plasma-liquid interfaces are notoriously nonlinear, multiscale in both space and time, and multiphysics**
- **The MOOSE finite element framework was selected as an appropriate platform for development of a general plasma software package:**
  - MOOSE applications are natively parallelizable and intended for high performance computing (HPC)
  - All MOOSE apps are able to be coupled together, facilitating multiphysics simulations
- **The MOOSE app Zapdos[1] was developed specifically for modeling plasma transport in 2015-2016**
  - As of 2017, only included support for electron and argon discharges
- **No chemistry capabilities were included in the MOOSE framework, and Zapdos was hard-coded to accept only a handful of reactions**



Electron density as a function of interfacial loss coefficient in the gas phase (left) and water phase (right). Simulation was performed with Zapdos. Figure adapted from [4].

[4] Lindsay, A. *et al.* J. Phys. D: Appl. Phys. *49* (2016) 235204 (9pp)

[5] C. DeChant, S. Keniley, D. Curreli, K. Stapelmann, S. Shannon, "Multi-physics simulation of the COST APPJ in the MOOSE framework", Bull. Am. Phys. Soc. 71th Annual Gaseous Electronic Conference, GT1.74, Portland, Oregon, Nov 5-9, 2018

Model Development

- **As of 2017, Zapdos was hard-coded to accept only four species (e^-, Ar^+ in the gas phase, and $e^-_{(aq)}$ and $OH^-_{(aq)}$ in the water), with 5 total reactions.**
- **As part of past NSF research, we introduced two new capabilities:**

1. Developed Plasma Chemistry Application in MOOSE: "CRANE"

https://github.com/lcpp-org/crane

- Written a model capable of handling an arbitrary number of reactions
- Reactions can be automatically parsed by code into source and sink terms
- Coupled to Zapdos to add source terms to drift-diffusion equations

2. Upgraded Zapdos

https://github.com/shannon-lab/zapdos

- Allowed an arbitrary number of user-defined species
- Included surface charge accumulation
- Upgraded water model to include neutral transport across interface

Zapdos: Drift-Diffusion-Reaction Equations

Volumetric Terms:

Species Density:

$$\frac{\partial n_s}{\partial t} + \nabla \cdot \vec{\Gamma}_s = \boxed{R_{sr}}$$

Electron Energy:

$$\frac{\partial (n_e \epsilon)}{\partial t} + \nabla \cdot \vec{\Gamma}_\epsilon = \underbrace{-e\vec{\Gamma}_e \cdot \vec{E}}_{\textit{Joule Heating}} + \boxed{R_{sj,\epsilon}}$$

$$\vec{\Gamma}_s = \pm \mu_s \vec{E} n_s - D_s \nabla n_s$$

$$\vec{\Gamma}_\epsilon = -\frac{5}{3}\epsilon \vec{\Gamma}_e - \frac{5}{3} n_e D_e \nabla \epsilon$$

Poisson Equation:

$$-\nabla^2 \phi = \frac{(\sum_i q_i n_i + q_e n_e)}{\epsilon_0}$$

Boundary Conditions [6]:

Electron BC:

$$\vec{\Gamma}_e \cdot \hat{n} = \frac{1-r_e}{1+r_e}\left[-(2a_e - 1)\mu_e \vec{E} \cdot \hat{n} n_e + \frac{1}{2} v_{th,e} n_e - \frac{1}{2} v_{th,e} n_\gamma\right] \cdot$$
$$- \frac{1}{2} v_{th,e} n_\gamma\big] - \frac{2}{1+r_e}(1 - a_e) \sum_i \gamma_i \vec{\Gamma}_i \cdot \hat{n}$$

Ion/Netural BC:

$$\vec{\Gamma}_i \cdot \hat{n} = \frac{1-r_i}{1+r_i}\left[\pm(2a_i - 1)\mu_i \vec{E} \cdot \hat{n} n_i + \frac{1}{2} v_{th,i} n_i\right]$$

Reaction Rates:

$$R_{sj} = \sum_j \nu_{sj} k_j \prod_r^R n_r$$

$$R_{sj,\epsilon} = \sum_j \nu_{sj} k_j \prod_r^R n_r \Delta\epsilon_j$$

[6] Hagelaar, G. *et al.* Physical Review E. *62*, 1 (2000)

CRANE: Chemical Kinetics

- **Crane is a standalone Moose application developed as part of the previous NSF work focused on modeling arbitrary systems of ODEs**
- **Source code:** https://github.com/lcpp-org/crane
- **When coupled to Zapdos, it provides the reaction rate portion of the drift-diffusion-reaction system**

$$\frac{dn_s}{dt} = \sum_{r=1}^{r_{max}} K_{sr}$$

$$K_{sr} = \underbrace{\nu_{sr}}_{\text{Stoichiometric Coefficient}} \underbrace{k_r}_{\text{Rate Coefficient}} \underbrace{\prod_l n_l^L}_{\text{Product of all Reactants for reaction } r}$$

- **Electron-impact reactions preprocessed with external Boltzmann solver (Bolsig+)**
  - Integral of EEDF $k_r = \gamma \int_0^\infty \varepsilon \sigma_r f_0 d\varepsilon$
  - Calculates rate coefficients (k) and electron transport coefficients
  - Values stored in look-up tables for a range of mean electron energies

- **Developed to allow an arbitrary number of reactions to be added in a human-readable format**

Reaction	Rate Coefficient	Units
$e + Ar \rightarrow e + Ar$	EEDF	$m^3\ mol^{-1}\ s^{-1}$
$e + Ar \rightarrow Ars + e$	EEDF	
$e + Ars \rightarrow e + Ar$	EEDF	
$e + Ar \rightarrow 2e + Ar^+$	EEDF	
$e + Ars \rightarrow 2e + Ar^+$	EEDF	
$Ars + Ars \rightarrow e + Ar + Ar^+$	3.3734×10^8	
$Ars + Ar \rightarrow Ar + Ar$	1.807×10^3	

Typical reaction list you find in a paper

How you write it in CRANE:

```
[Reactions]
  [argon_reactions]
    species = 'em Ar+ Ar*'
    file_location = 'rate_files'
    potential = 'potential'


    reactions = 'em + Ar -> em + Ar                    : EEDF [elastic]  (reaction1)
                 em + Ar -> em + Ar*                   : EEDF [-11.5]    (reaction2)
                 em + Ar* -> em + Ar                   : EEDF [11.5]     (reaction4)
                 em + Ar -> em + em + Arp              : EEDF [-15.76]   (reaction3)
                 em + Ar* -> em + em + Arp             : EEDF [-4.43]    (reaction5)
                 Ar* + Ar* -> em + Ar + Arp            : 3.3734e8'
                 Ar* + Ar -> Ar + Ar                   : 1807
  []
[]
```

[7] S. Keniley, D. Curreli, C. DeChant, S. Shannon, CRANE: a Novel MOOSE-based Open-Source tool for Plasma Chemistry Applications and Code Coupling, Bull. Am. Phys. Soc., GT1.39, Portland, Oregon, Nov 5-9, 2018

2. Upgrades of Zapdos

Source code: https://github.com/shannon-lab/zapdos

Zapdos required multiple updates to address realistic plasma-water chemistry:

2.1 Accept arbitrary number s of user-defined plasma species

2.2 Add surface charge accumulation for dielectric interfaces

2.3 Include heavy species solvation and evaporation boundary conditions

$$\frac{\partial n_s}{\partial t} + \nabla \cdot \vec{\Gamma}_s = R_{sr}$$

$$\vec{\Gamma}_s = \pm \mu_s \vec{E} n_s - D_s \nabla n_s$$

$$-\nabla^2 \phi = \frac{(\sum_i q_i n_i + q_e n_e)}{\epsilon_0}$$

2. Upgrades of Zapdos

2.1 Accept arbitrary number of user-defined species

- Existing code was abstracted to include arbitrary species variables
- A new class, ‘**HeavySpeciesMaterial’,** was added to add species properties (mass, charge, transport coefficients)
- Mobility and diffusivity are by default given by Einstein’s relation (user can change)

```
[gas_species_example]
  type = HeavySpeciesMaterial
  heavy_species_name = Ar+
  heavy_species_mass = 6.64e-26
  heavy_species_charge = 1.0
  diffusivity = 1.6897e-5
[]
```

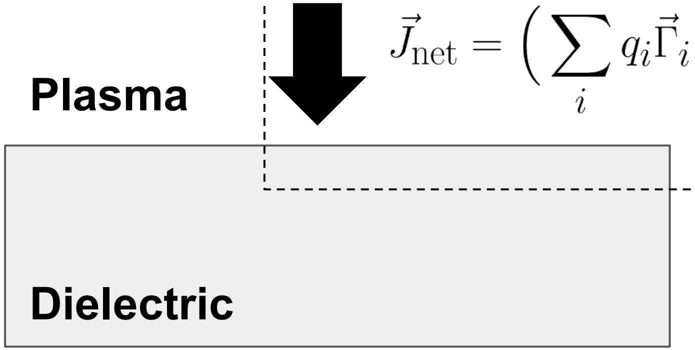
$$\mu_s = \frac{Z_s q_e D_s}{k_B T_e}$$

2. Upgrades of Zapdos

2.2 Added surface charge accumulation for dielectric interfaces

- Dielectrics are widely used in plasma discharges, but no interface existed in Zapdos to handle surface charge accumulation

- Surface charge was added to the model in two parts:
  a. ODE at dielectric boundary to describe surface charge accumulation
  b. Interfacial boundary condition for discontinuous electric field



$$\vec{J}_{\text{net}} = \left(\sum_i q_i \vec{\Gamma}_i - q_e \vec{\Gamma}_e \right) \cdot \hat{n}$$

$$\frac{d\sigma}{dt} = \vec{J}_{\text{net}}$$

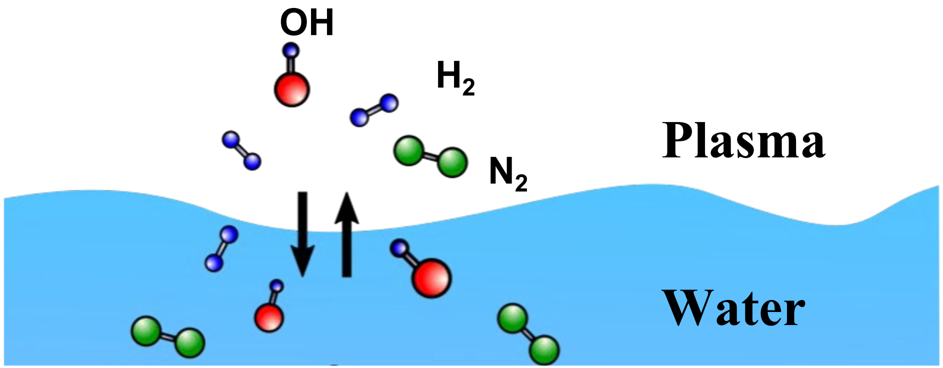
$$(\epsilon_p \nabla \phi_p - \epsilon_d \nabla \phi_d) \cdot \hat{n} = \sigma$$

2. Upgrades of Zapdos

2.3 Include heavy species solvation and evaporation boundary conditions

- A two-way interfacial transport model was added to Zapdos to allow neutral species to transport between gas and liquid phases based on Henry's law
  a. Henry coefficient, H, defines equilibrium concentration of species at interface
  b. Flux equality at the interface allows species to naturally flow in or out of the liquid

- While Henry's law is an equilibrium relationship, but only a ***local*** equilibrium at the interface is assumed - no assumption about bulk concentrations is made



Henry's Law (local at the interface):

$$H n_G = n_L$$

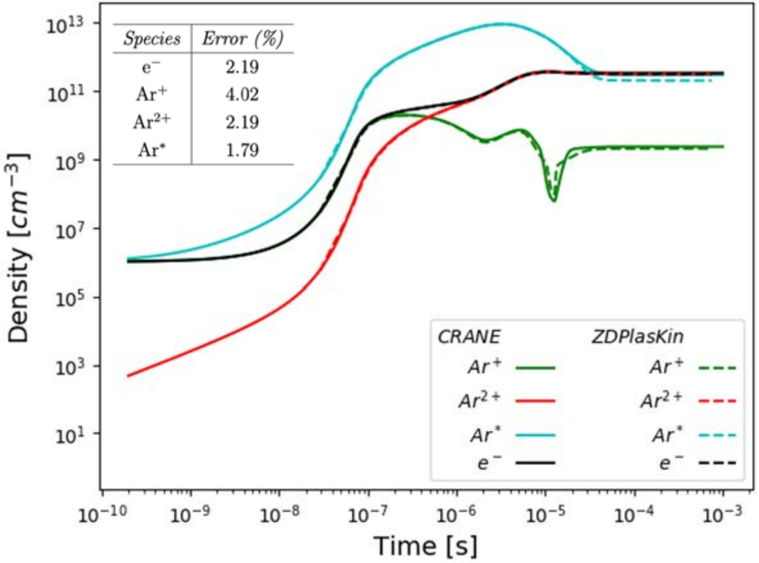
Flux Equality:

$$D_G \nabla n_G = D_L \nabla n_L$$

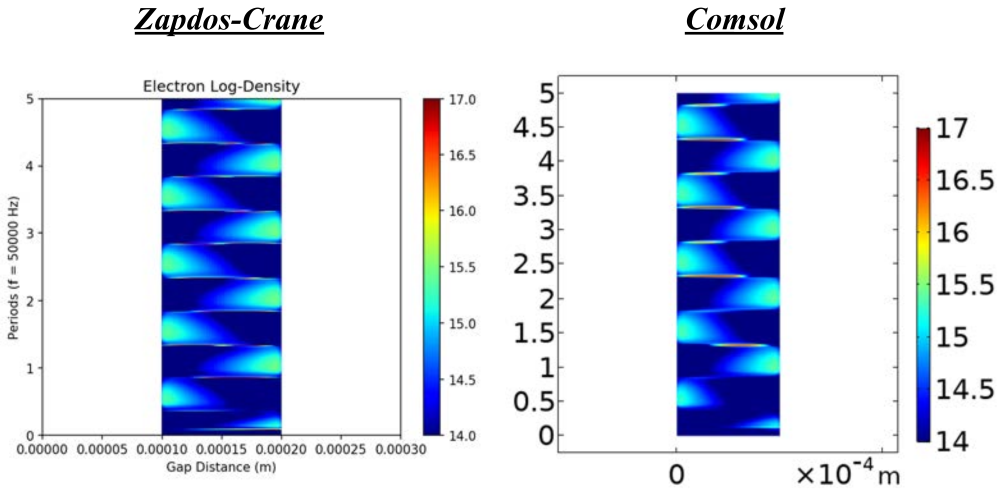
Verification of Zapdos-Crane

- **Both codes were verified against multiple known problems; two examples:**

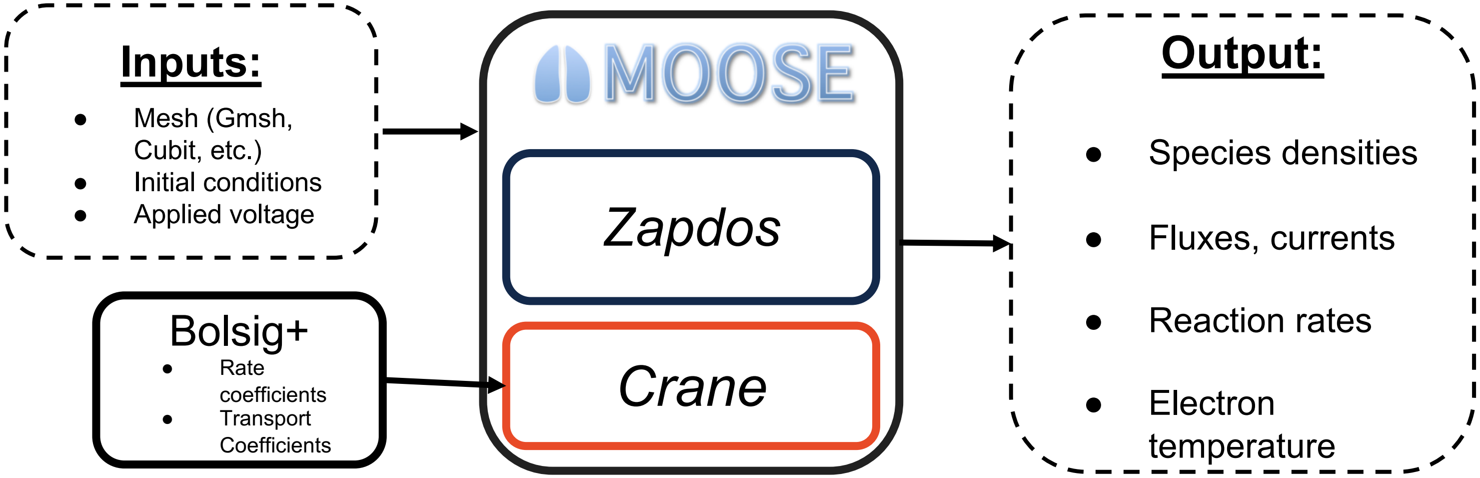
Crane vs. ZDPlasKin
(0D reaction networks)



Zapdos-Crane vs. Comsol
(1D Dielectric Barrier Discharge)



Typical Workflow



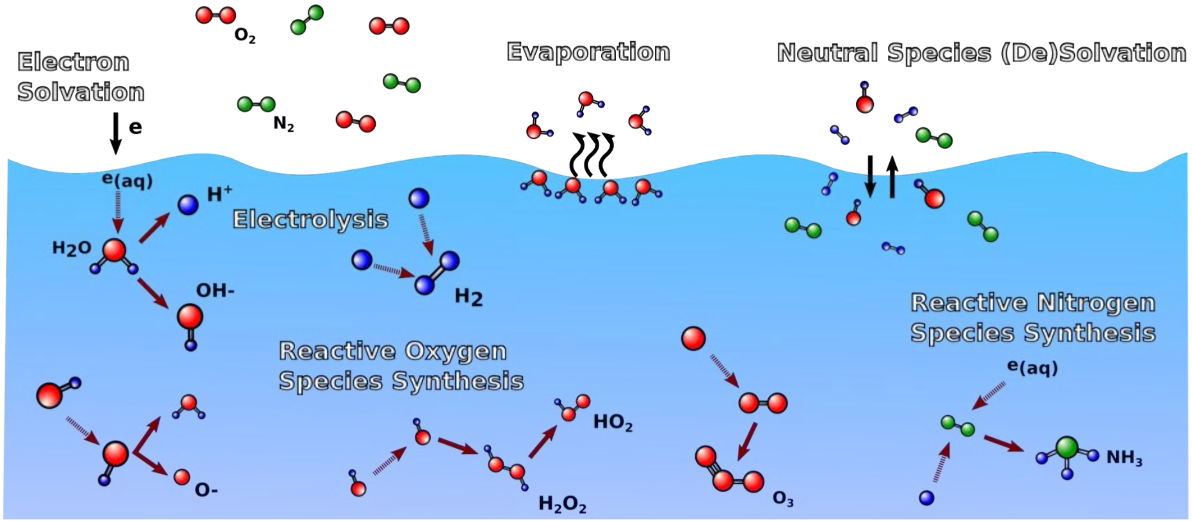
Zapdos-Crane was presented at a 2018 APS-GEC Workshop as an open-source plasma tool :

[8] C. Icenhour, S. Keniley, C. DeChant, C. Permann, A. Lindsay, R. Martineau, D. Curreli, S. Shannon, Multi-Physics Object Oriented Simulation Environment (MOOSE), Bull. Am. Phys. Soc. 71th Annual Gaseous Electronic Conference, BM2.1, Portland, Oregon, Nov 5-9, 2018

https://github.com/lcpp-org/crane

https://github.com/shannon-lab/zapdos

Plasma-liquid interfaces: a challenge for modern plasma



- **Multiscale and multiphysics**
  - Electron penetration depth: ~10-100 nm
  - Discharges: mm-m
  - Electron solvation: O(fs)
  - Electron-driven aqueous reactions: O(ns)
  - Chemical reactions: O(us-ms)
  - Species diffusion: O(ms - minutes)
- **Strongly coupled behavior between plasma and water**
  - Electrons drive chemistry in the interface layer, which change chemical composition of the water
  - Species diffuse in and evaporate out of interface, modifying plasma discharge conditions
  - Electric fields, gas flow can deform water
  - Plasma-induced fluid convection and turbulence is possible

Model of the Plasma-Water Interface in Zapdos-Crane

- **Water region assumed to behave as a "dense plasma":**
  - Same drift-diffusion-reaction equations apply
  - Higher background density
  - Relative permittivity of 81

Assumptions:

- **Electrons solvate instantly in water phase**
  - Solvation time estimated to be O(fs)
- **Heat transport is neglected (recently relaxed)**
- **Electron temperature is not considered in water**

Plasma Region:

$$\frac{\partial n_s}{\partial t} + \nabla \cdot \vec{\Gamma}_s = R_{sr}$$

$$-\nabla^2 \phi = \frac{(\sum_i q_i n_i + q_e n_e)}{\epsilon_0}$$

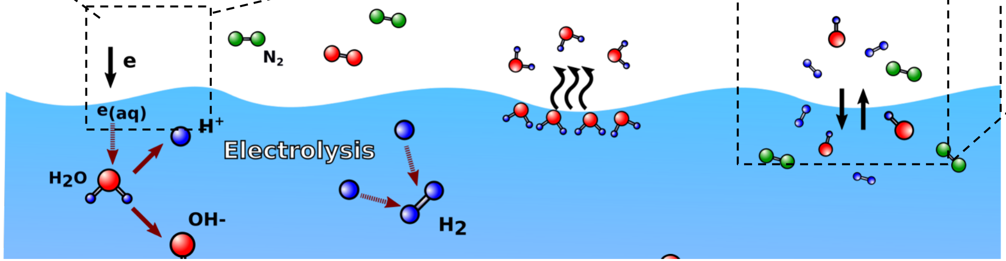
Water Region:

$$\frac{\partial n_{s,aq}}{\partial t} + \nabla \cdot \vec{\Gamma}_{s,aq} = R_{sr,aq}$$

$$-\nabla^2 \phi = \frac{(\sum_i q_i n_i + q_e n_e)}{\epsilon_0}$$

Electrons directly drift and diffuse into water:

$$\vec{\Gamma}_{e,\text{liquid}} \cdot \hat{n} = -\vec{\Gamma}_{e,\text{gas}} \cdot \hat{n}$$



Heavy Species Solvation (Henry's Law):

$$H n_G = n_L$$

$$D_G \nabla n_G = D_L \nabla n_L$$

[10] Shane Keniley, Davide Curreli, Corey DeChant, and Steve Shannon, Numerical Modeling of the Plasma-Liquid Interface using the Zapdos-CRANE Open-Source Package, 72nd Annual Gaseous Electronics Conference, College Station, Texas, October 28-November 1, 2019

Case 1: Ar/H_2O plasma on liquid water

A first test of plasma-water interactions was developed in humid argon at atmospheric pressure

- DC circuit with ballast resistor boundary condition was applied
- Grounded wall at x = 1.01 mm
- Plasma-water interface at x = 1 mm

Left: pin-to-water discharge. (Adapted from [4])

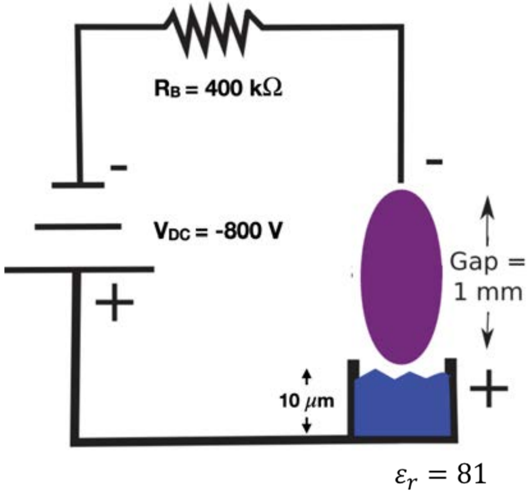
Bottom: Simulation domain

Plasma model:

- 26 species, 141 reactions
- Humid Argon
- Neutral Ar included as static background
- Reaction network largely adapted from Tian and Kushner [11]

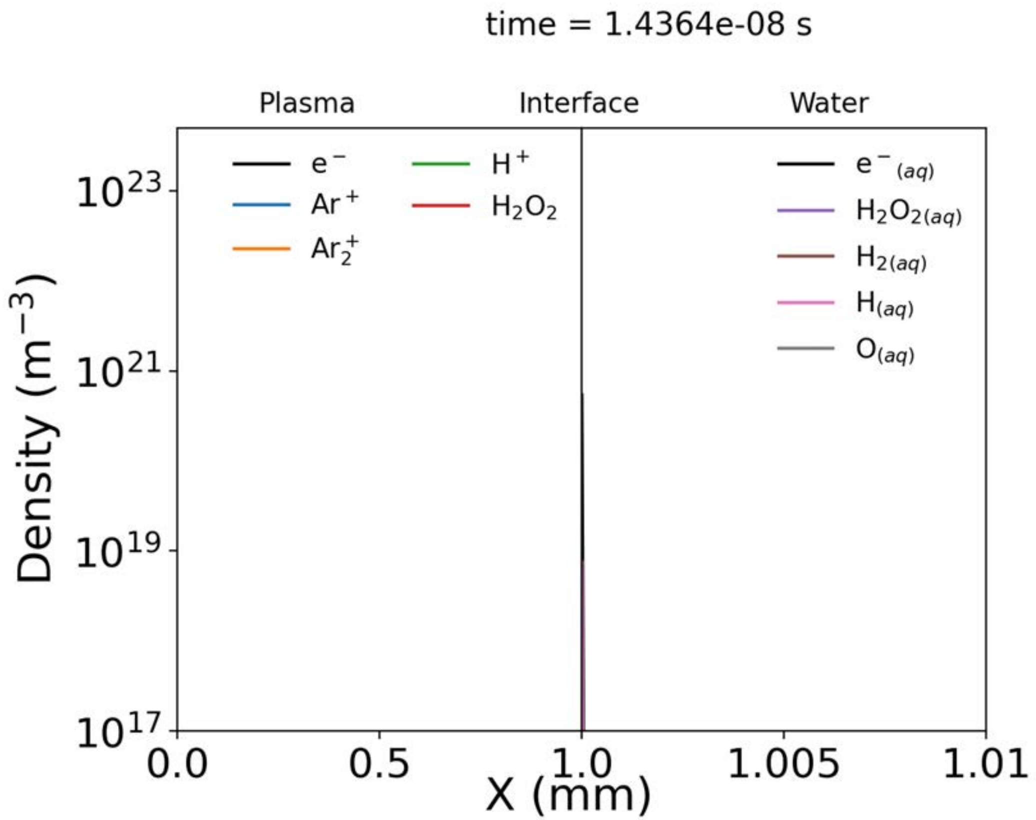
Water model:

- 17 species, 67 reactions
- H_2O static background
- Salt included at initial concentration of 10 mM
  - Artificial source term is used to simulate replenishment from bulk for Na^+ and Cl^-

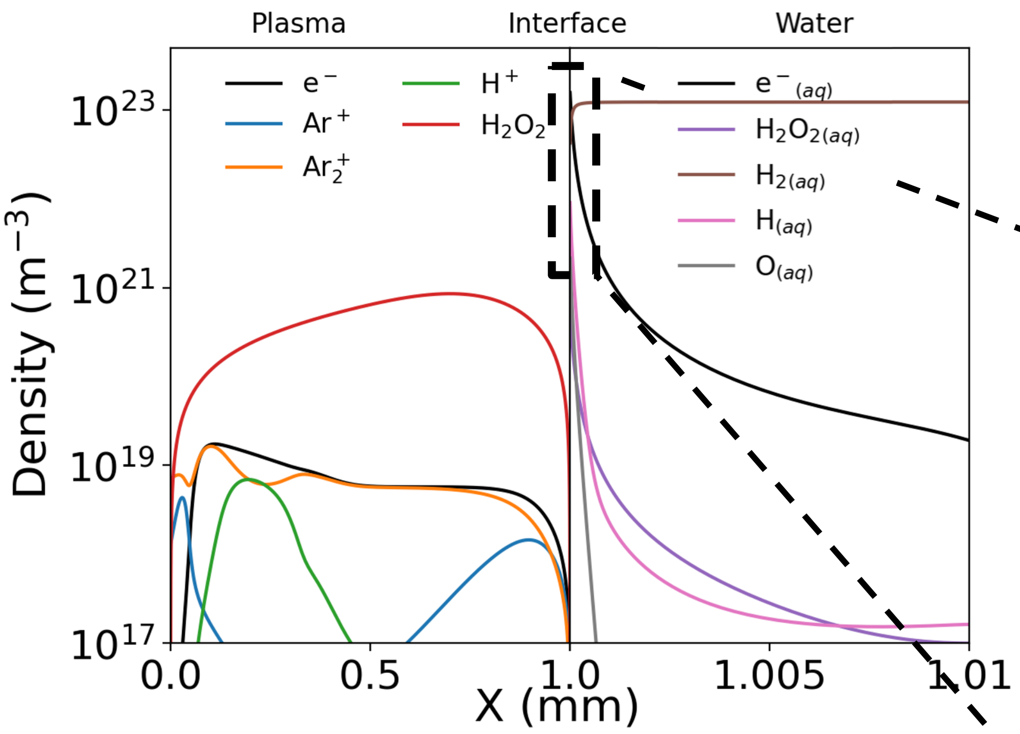


[4] Lindsay, A. *et al.* J. Phys. D: Appl. Phys. *48* (2015) 424007, [11] Tian, W. Phd Dissertation, University of Michigan, 2015.

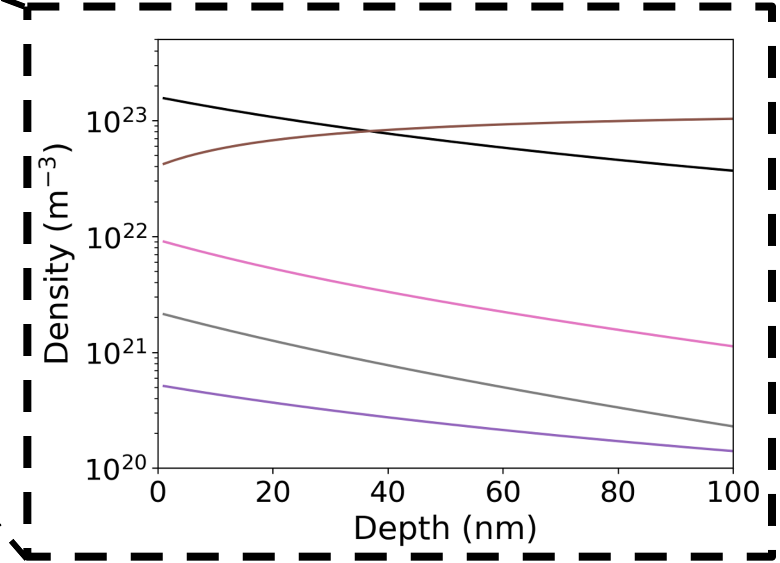
Results - Ar/H_2O plasma on liquid water, species density



Results - Ar/H_2O plasma on liquid water, species density



- **Electrons reach high concentrations on the liquid side**
- **They react quickly, decreasing by a factor of 5 within the first 100 nm**



Results - Aqueous Charge Balancing

- **Electrons quickly convert to OH^- through second-order recombination:**

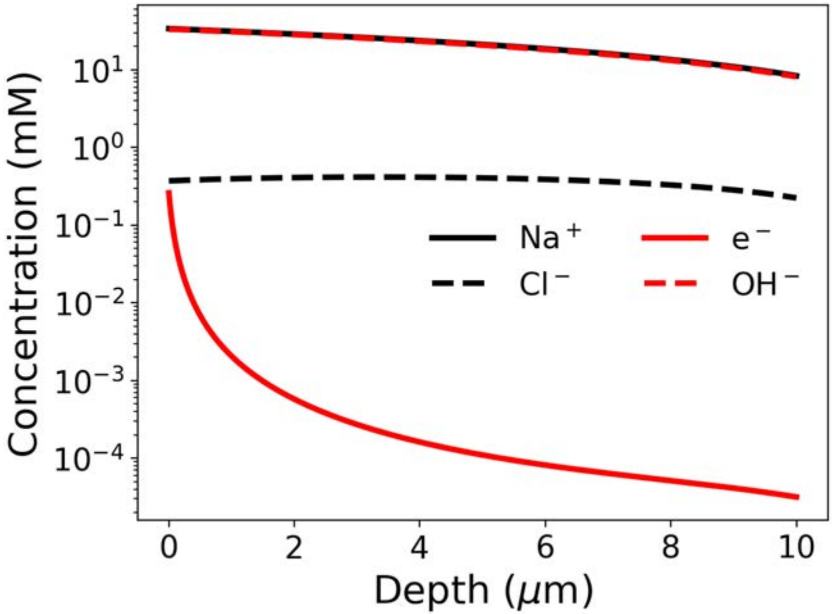
$$2e_{(aq)} + 2H_2O \rightarrow 2OH^-_{(aq)} + 2H_{2(aq)}$$

- **OH^- accumulation forms highly basic solution at the interface**

- **Na^+ accumulates at the surface to balance the negative charge injected by the plasma**

- **This effect was predicted by Rumbach [12] but not seen in previous models [13]**

1 mM = 1 mol m^{-3}



[12] Rumbach, P. *et al*. Physical Review E, 95(5):53203 (2017)
[13] Gopalakrishnan, R. *et al*. J. Phys. D: Appl. Phys. 49 29 (2016)

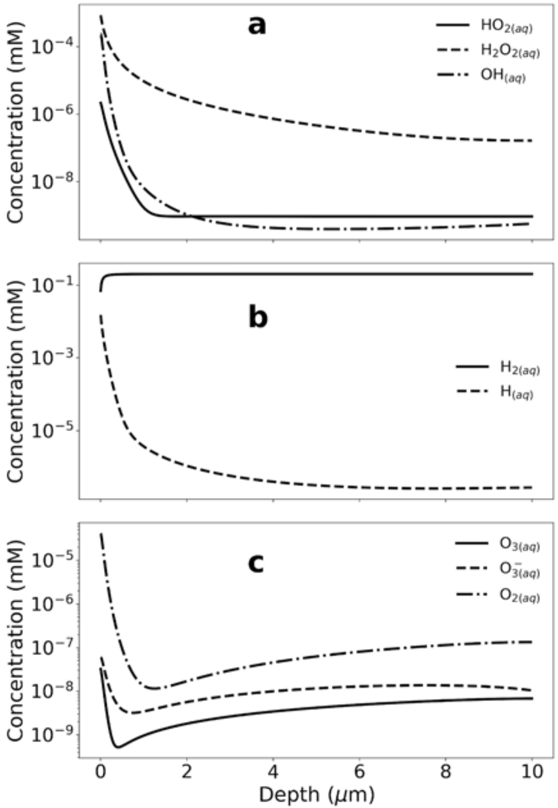
Results - Reactive Species Generation

- **HO_2, H_2O_2, and OH are the most dominant electron scavengers via reactions:**

$$e_{(aq)} + HO_2 \rightarrow HO_2^-$$
$$e_{(aq)} + H_2O_{2(aq)} \rightarrow OH_{(aq)} + OH^-_{(aq)}$$
$$e_{(aq)} + OH_{(aq)} \rightarrow OH^-_{(aq)}$$

- **Hydrogen ($H_{2(aq)}$) reaches high concentrations in the liquid, as expected from electrolysis**

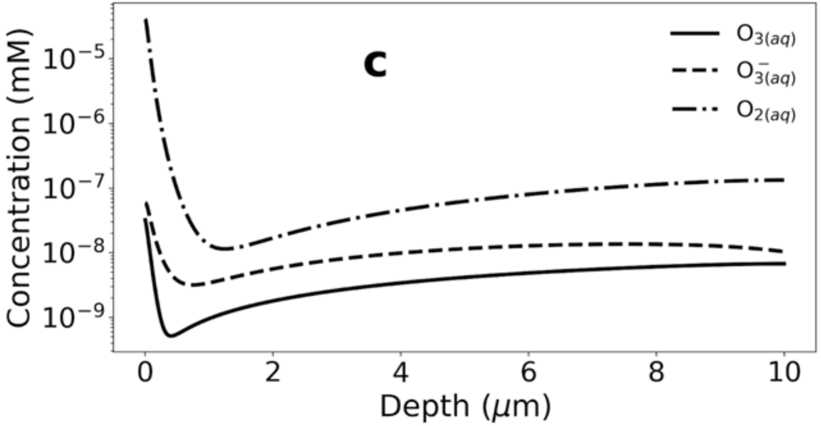
- **Oxidizing species (O_2, O_3) are quickly depleted in the interface layer by solvated electrons**
    - Increasing solvated electron concentration in the liquid phase should *decrease* the concentration of oxidizing agents in the liquid



Solvated Electrons at the Water Interface

I. Aqueous Chemical Pathways

- **Solvated electrons have been shown to play a dominant role in the plasma-water interface**
- **While solvated electrons clearly cause a decrease in oxygen species in the thin interface region (< 1 micron), they also open up additional pathways in the water**



Based on the model results we expect that: Increasing electron current density will *decrease* delivery of O_3 and H_2O_2 due to aqueous chemical reaction pathways with solvated electrons.

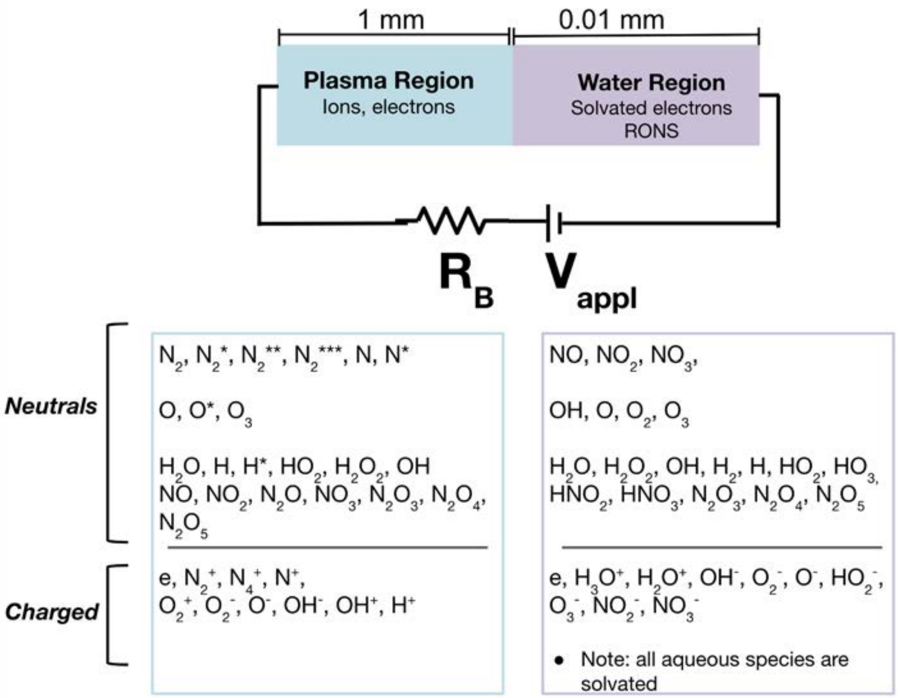
$$e_{(\mathrm{aq})} + O_{3(\mathrm{aq})} \rightarrow O^-_{3(\mathrm{aq})}$$

$$e_{(\mathrm{aq})} + O^-_{3(\mathrm{aq})} \rightarrow O_{2(\mathrm{aq})} + 2OH^-_{(\mathrm{aq})}$$

$$e_{(\mathrm{aq})} + H_2O_{2(\mathrm{aq})} \rightarrow OH_{(\mathrm{aq})} + OH^-_{(\mathrm{aq})}$$

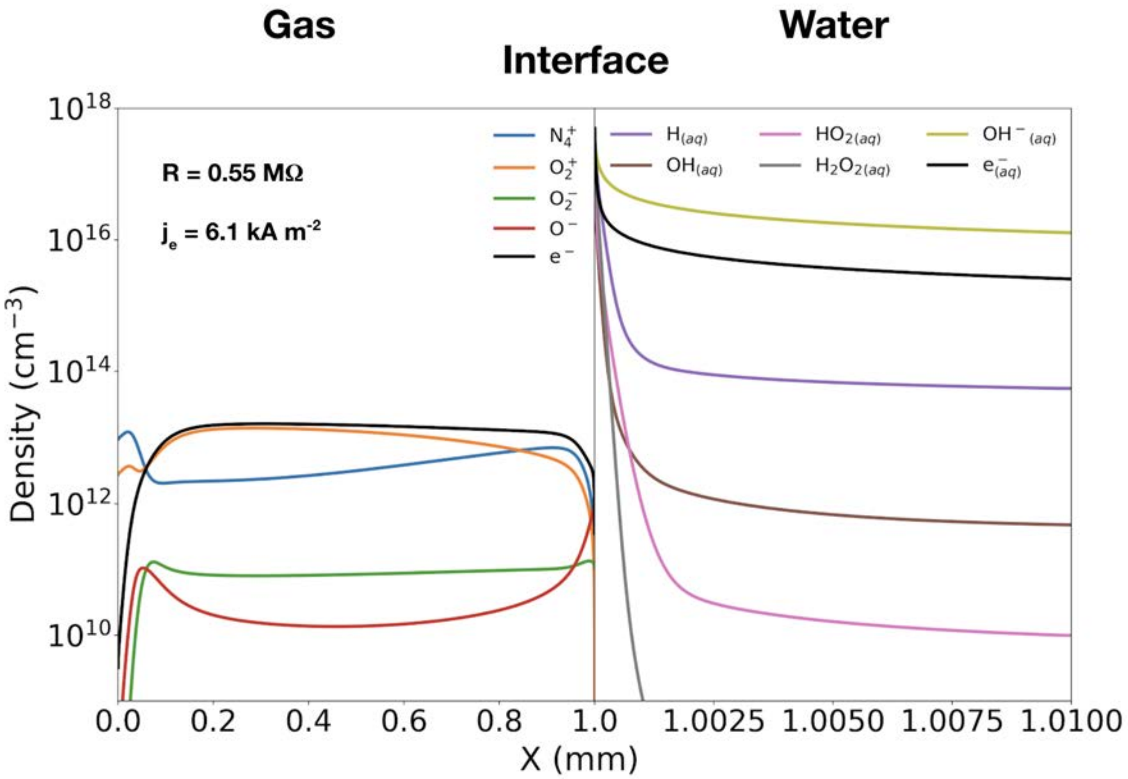
Case 2: Air/H_2O (humid air) plasma on liquid water

- **This case involves an atmospheric pressure mixture of nitrogen, oxygen, and water**
  - 1D DC discharge, two region model
  - Grounded wall at liquid boundary (x = 1.01 mm)
  - V_{appl} = -3 kV
  - Both air and liquid chemistry models are adapted from Tian's 2015 thesis and Buxton et al [4] [7]
- **Humid air plasma model (N_2/O_2/H_2O)**
  - 32 species, 187 reactions
  - Nitrogen, water, and hydrogen: Itikawa database
  - Oxygen: TRINITI database; both at: www.lxcat.net
  - Water introduced with vapor pressure BC at liquid interface (amounts to ~1% humidity)
- **Water model**
  - 29 species, 93 reactions
  - Permittivity: $\varepsilon_r = 81$
  - Initial pH of 7 ($[H_3O^+_{aq}]$ = $[OH^-_{aq}]$ = $5x10^{11}$ cm^{-3})
  - Solvation occurs instantly upon entering water phase

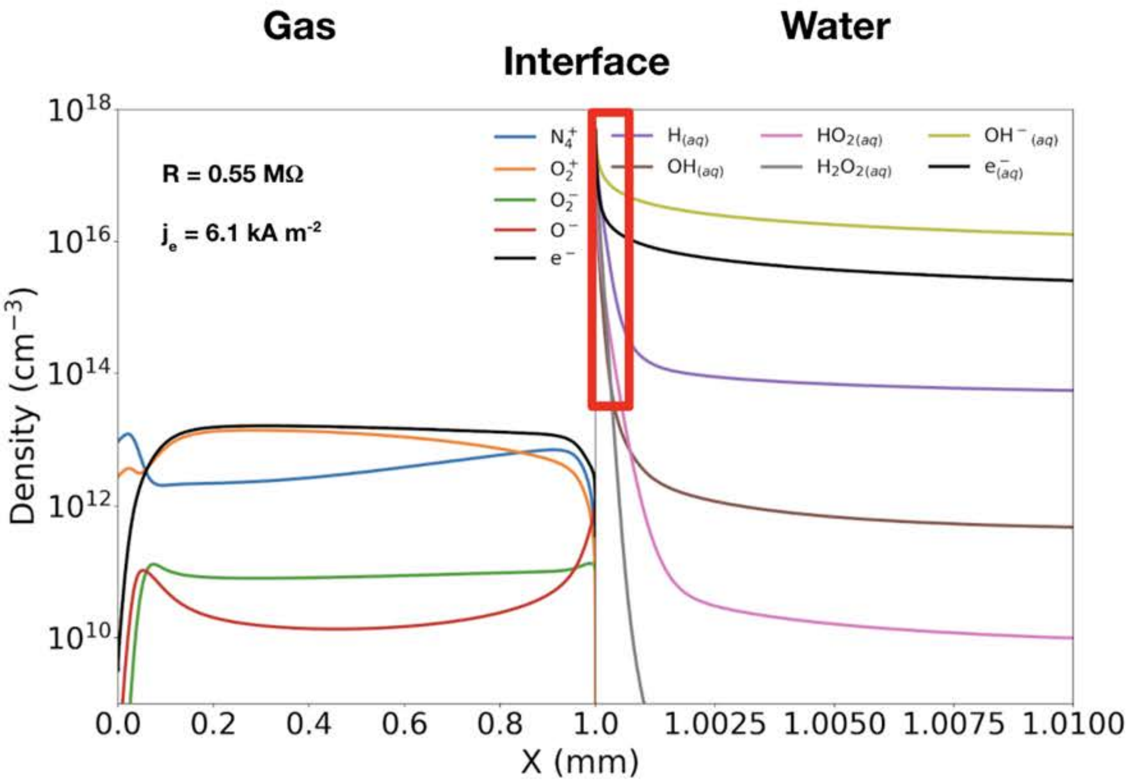


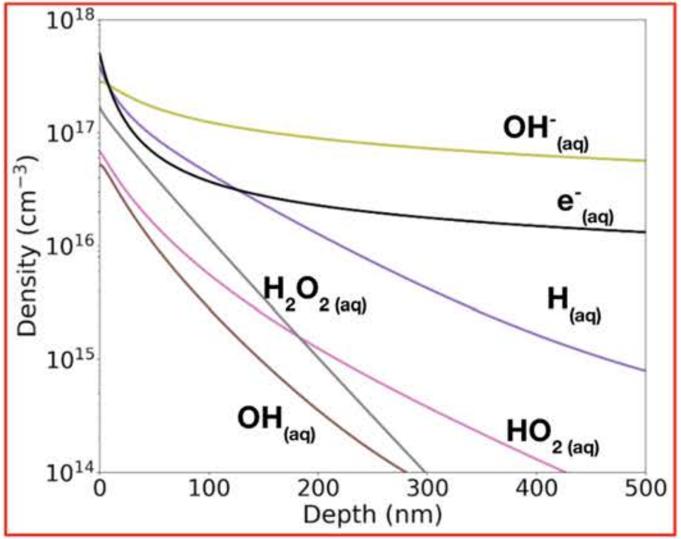
[4] Tian, Wei. "MODELLING INTERACTIONS OF ATMOSPHERIC PRESSURE PLASMAS AND WITH LIQUIDS" PhD Dissertation. University of Michigan (2015)
[7] Buxton, G. et al. "Critical review of rate constants for reactions of hydrated electrons, hydrogen atoms and hydroxyl radicals." Journal of Phys. Chem. Ref. Dat. 17 2 (1988)

Case 2: Air/H_2O (humid air) plasma on liquid water, density



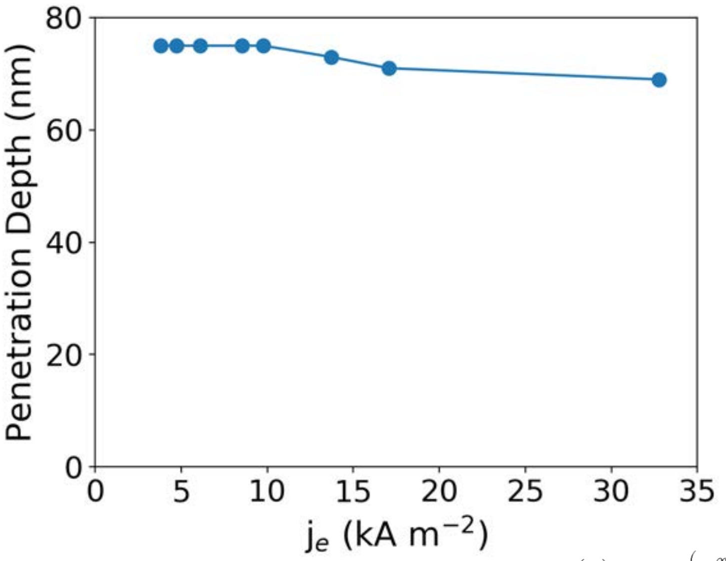
Case 2: Air/H_2O (humid air) plasma on liquid water, density

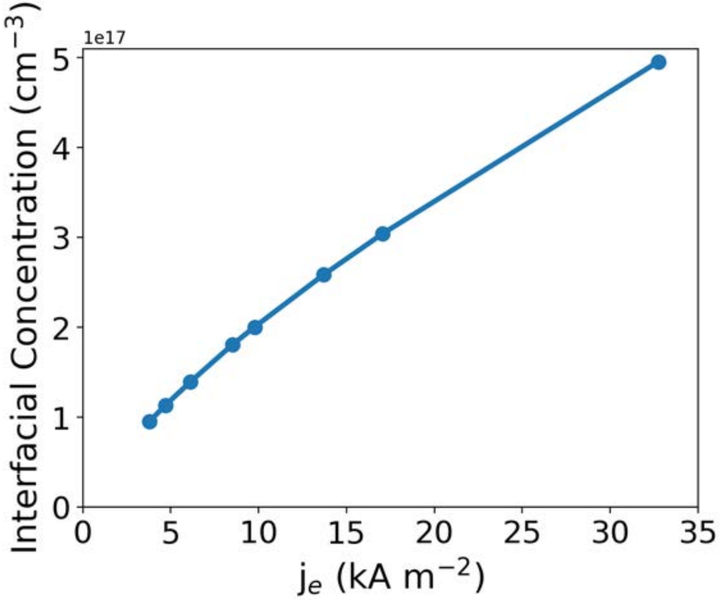




Solvated Electrons, Penetration Depth

- In all cases the solvated electrons quickly react within the first 100 nm
- Penetration depth* slightly decreases with electron current density
- 75 nm at 4 kA m^{-2} to 69 nm at 33 kA m^{-2}





*PD is here defined as 10% of the surface value, not classical "l" $n(x) = n_0 e^{(-x/l)}$

Solvated Electrons, Reaction Pathways

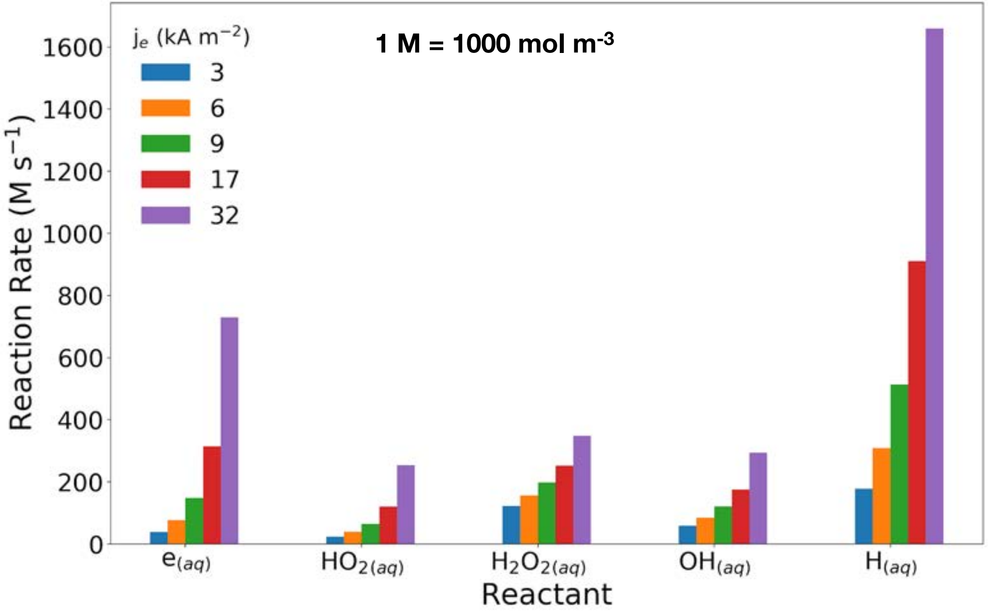
What is happening in the first 80 nm?

- The thin electron layer is very chemically active
- Electrons react significantly with all other radicals in the liquid phase
- Second order recombination rate increases quickly with electron concentration
- The terminal product of most reactions is hydroxide OH^-

$$e_{(aq)} + HO_{2(aq)} \rightarrow HO^-_{2(aq)}$$

$$e_{(aq)} + HO^-_{2(aq)} + H_2O \rightarrow OH_{(aq)} + 2OH^-_{(aq)}$$

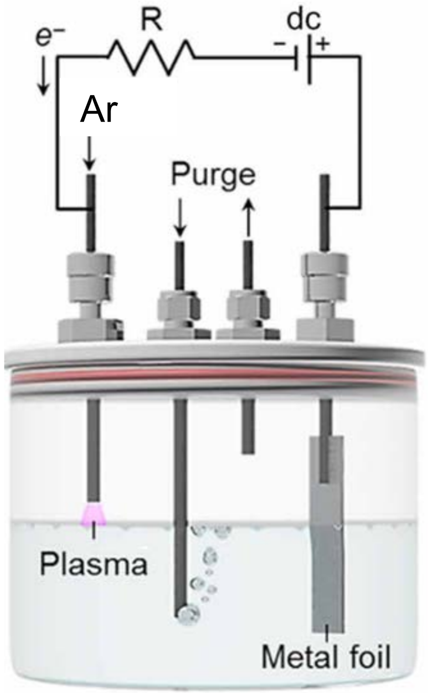
Dominant reactions with $e_{(aq)}$* averaged over first 80 nm



*Reaction also includes H_2O

Recently Started Validation of Plasma-Water Interaction Model

- The model presented in this work must be validated to ensure that it is accurately simulating plasma-liquid chemistry
- Experiments carried out by Prof. Sankaran's group (UIUC) are measure aqueous conditions (reactive oxygen species concentrations, pH) after plasma treatment
- The aqueous chemistry and plasma-water interaction model is compared to experimental measurements of aqueous species and pH
- Parametric scan of plasma parameters are performed to observe changes in reactive species production



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Conclusions

- **Crane and Zapdos are two new, open-source, software applications available to the LTP community, which can be used for the simulation of Low Temperature Plasmas with complex Plasma Chemistry**

- **Crane and Zapdos were used to study Plasma-water interactions, allowing innovative features, such as:**
  - Gas and liquid phases dynamically and implicitly coupled
  - Water affects discharge, and vice-versa

Future Work:

- **Experimental validation of plasma-water model**
  - Qualitative and semi-quantitative validation of the plasma-water model
  - Comparison to experimental measurements (Sankaran) of a plasma-water interface electrolytic cell

- **Tackle the challenge of solving the coupled nature of plasma-water interface**
  - Develop physically meaningful definition of solvated electron penetration depth, compatible with observations
  - Determine how solvated electrons affect the generation of selected reactive species in the water

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