



# Self-organization and Electrolyte Ion Mass Transport Processes with Chemistry in 1 ATM DC Glows

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# Abstract

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In plasma physics, self-organization is observed in phenomena ranging from plasmoid formation in low pressure, RF plasmas to large-scale, and magnetized structures observed on the surface of the sun. Of recent interest is the puzzling formation of self-organization patterns on the surface of liquid anodes in 1 ATM DC glows. While these patterns are of academic interest in regards to understanding collective phenomena, the appearance of the patterns may play an important role in the sub-surface liquid phase chemistry, driving convection and inducing thermal gradients. In this current work, a new, complex, star-shaped structure with round edges was observed with a copper sulfate electrolyte. The pattern was not observed with sodium chloride solutions. This observation suggests that electrolyte ion mass or perhaps ionization state may play a key role in determining overall pattern shape. In order to understand the role of the transport of electrolyte ions from liquid to the gas phase on discharge maintenance, and pattern formation, spectroscopic analysis of the halo surrounds the main plasma column for multiple electrolytes are studied as a function of discharge conditions.

*Keywords: DC atmospheric glows, plasma, self-organization, spectroscopic characteristic, electrolytes, chemistry .*

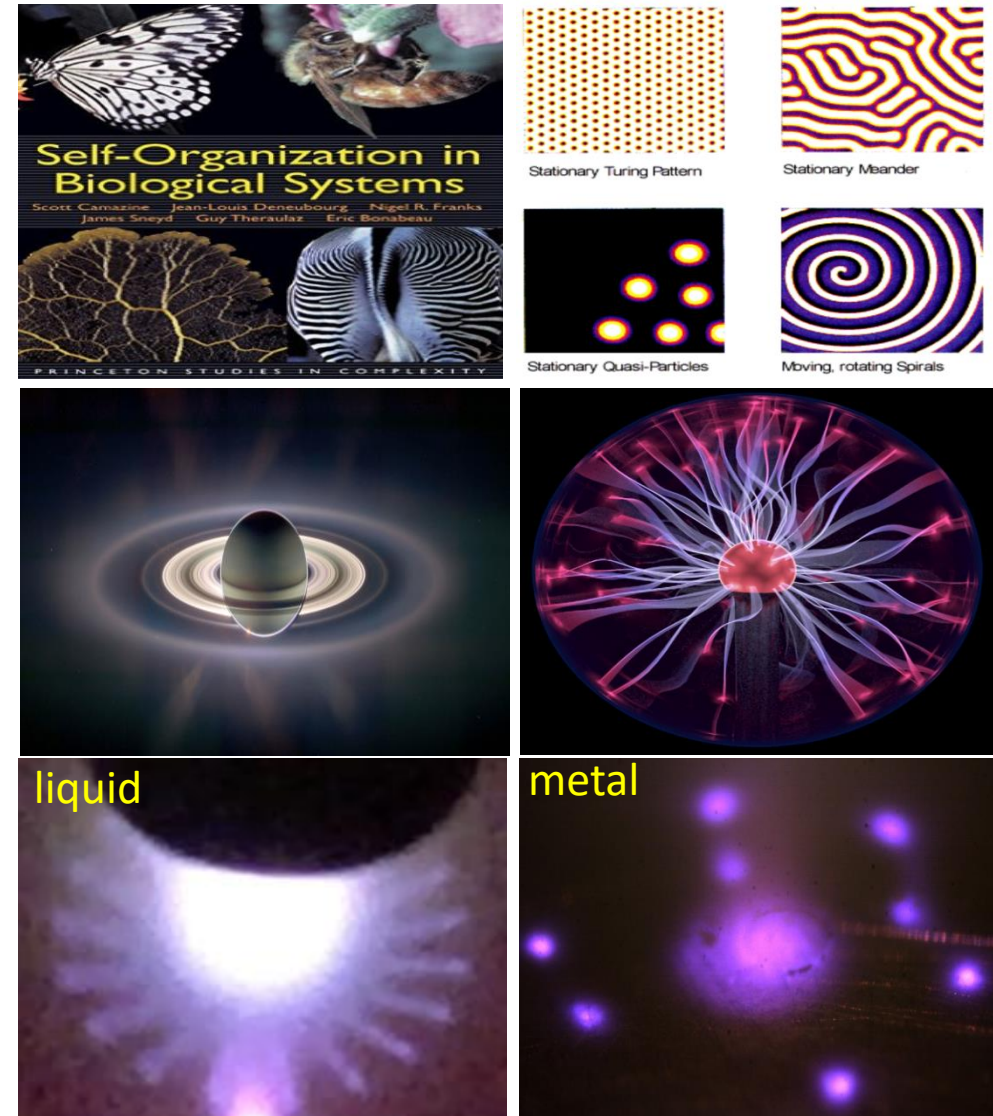
# Self-organization-Background

## Motivation

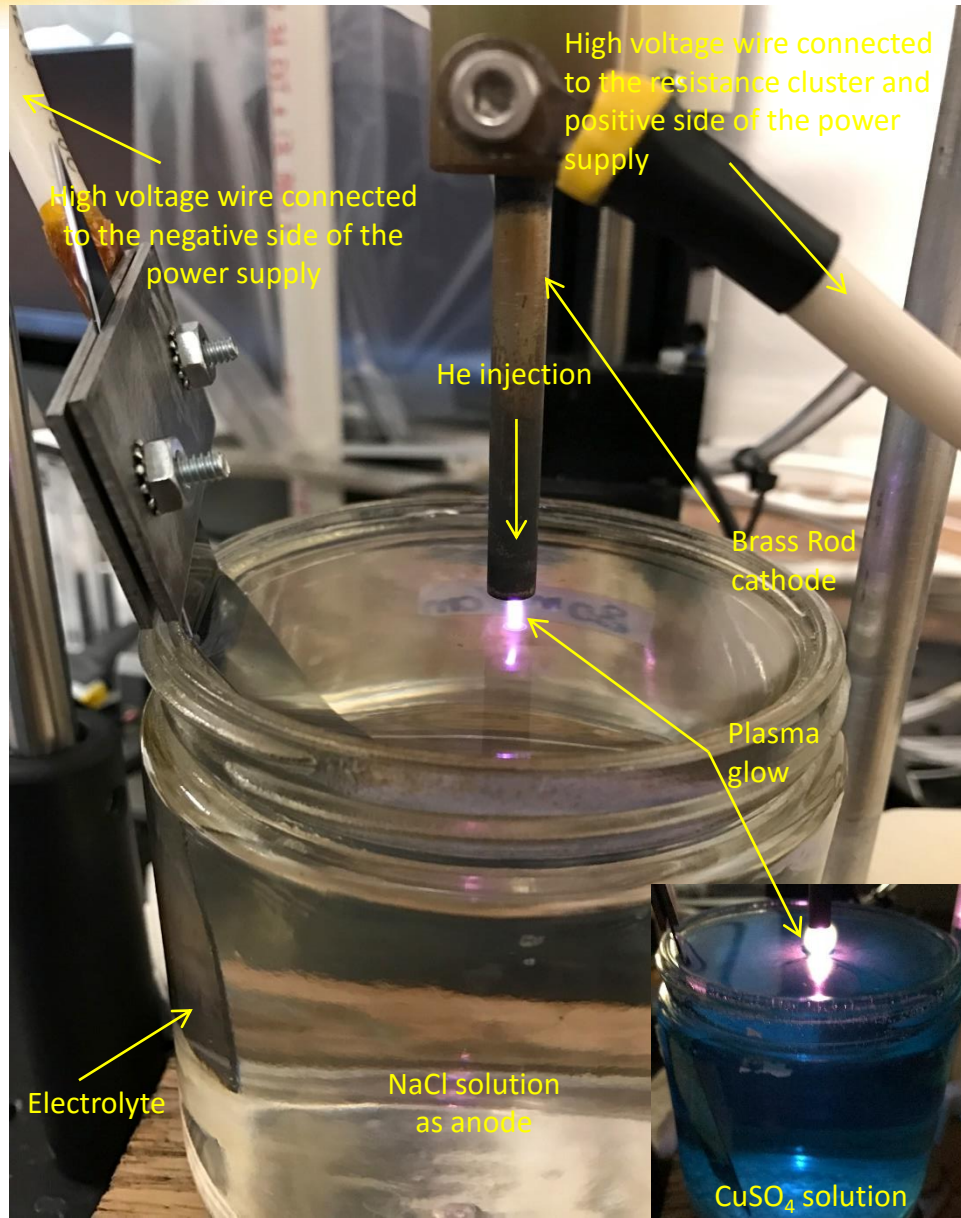
- Self-organization, is a process where some form of overall order arises from local interactions between parts of an initially disordered system.
- Self-organization patterns occurs in a variety of physical, chemical, biological, and cognitive systems.
- In plasma physics, self-organization is observed in phenomena ranging from plasmoid formation in low pressure, RF plasmas to large scale, and magnetized structures on the surfaced of the sun.

## Self organization in DC glows with liquid anode

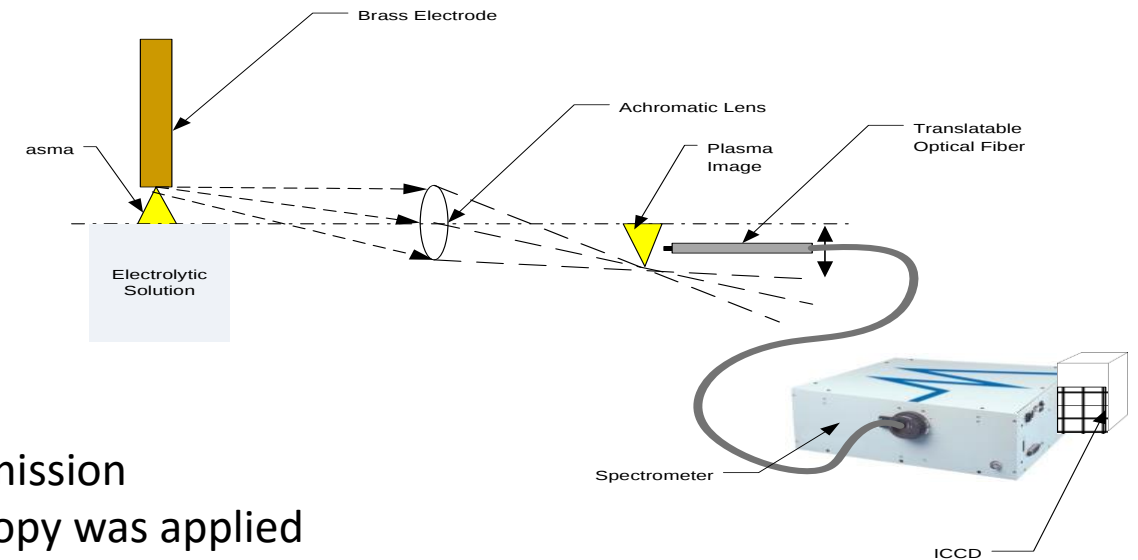
- Self-organization anode pattern observed on both liquid and metal surfaces
- Why do self-organized patterns form?
  - What role does anode fall region electric play?
  - What is the composition and plasma properties of patterns?
  - What is the relationship between the shape and motion of the patterns with current continuity at interface?



# Experimental Setup with Operating Condition

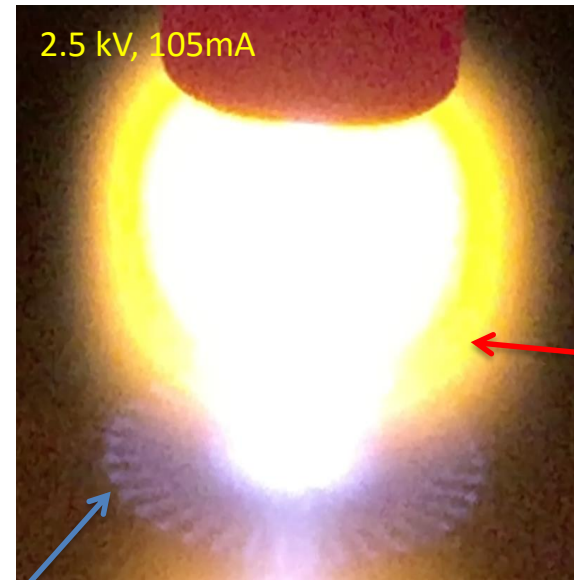
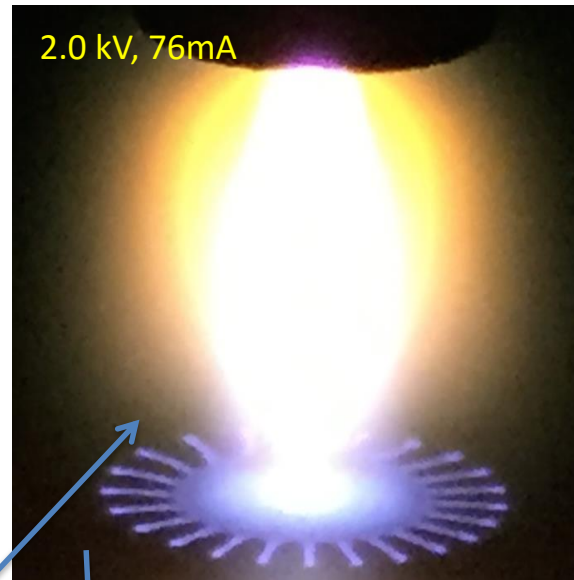
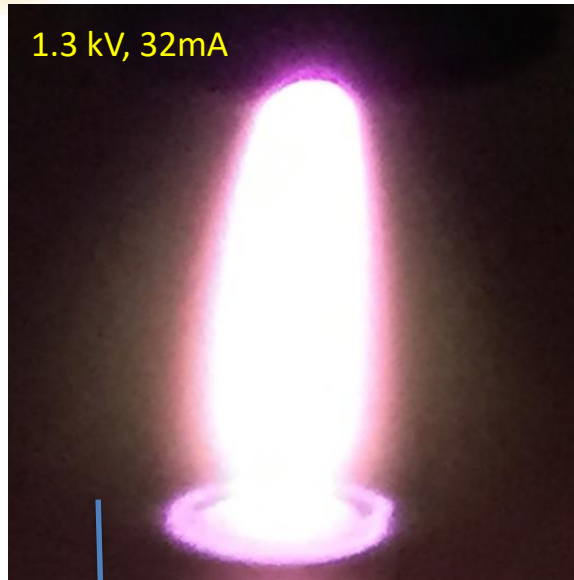


- ◆ He gas flow: 200 sccm; Applied Voltage range: 0-2.5kV; Current range: 0-120mA with gap controlled up to 8mm.
- ◆ Both NaCl solution (mainly focused) and  $\text{CuSO}_4$  solution (Cupric Fulfate + DI water) with initial conductivity of 12.00 mS/cm were tested in voltage controlling mode.
- ◆ Multiple electrolytes were tested with the same concentration of 8.735g/L in constant current control mode.

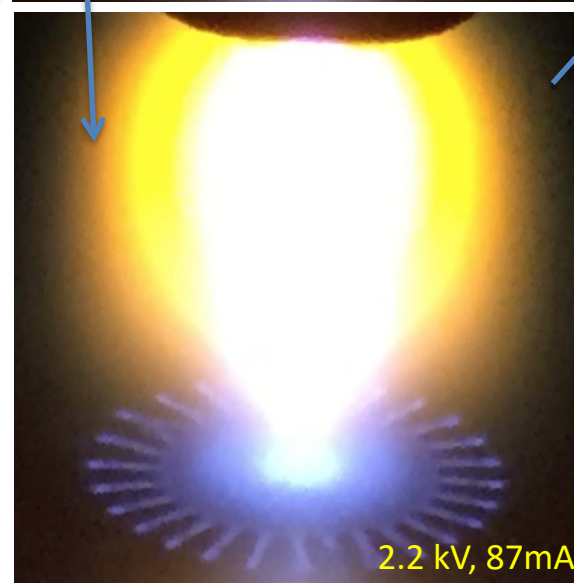
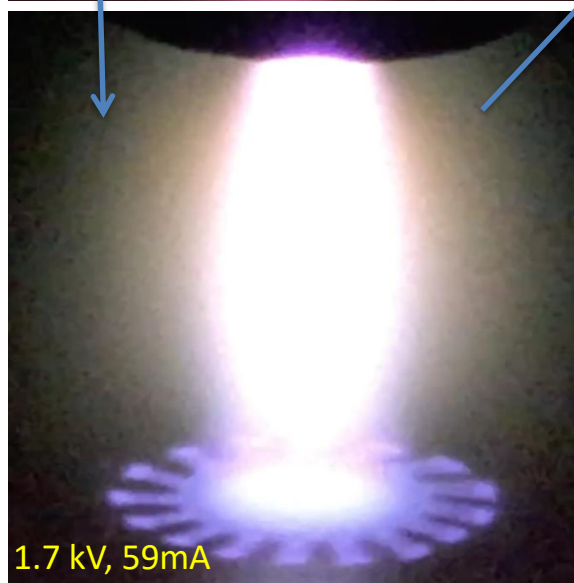


- ◆ Optical emission spectroscopy was applied as diagnostics.

# NaCl solution I: Self-organization with He Plasma Injection-liquid anode



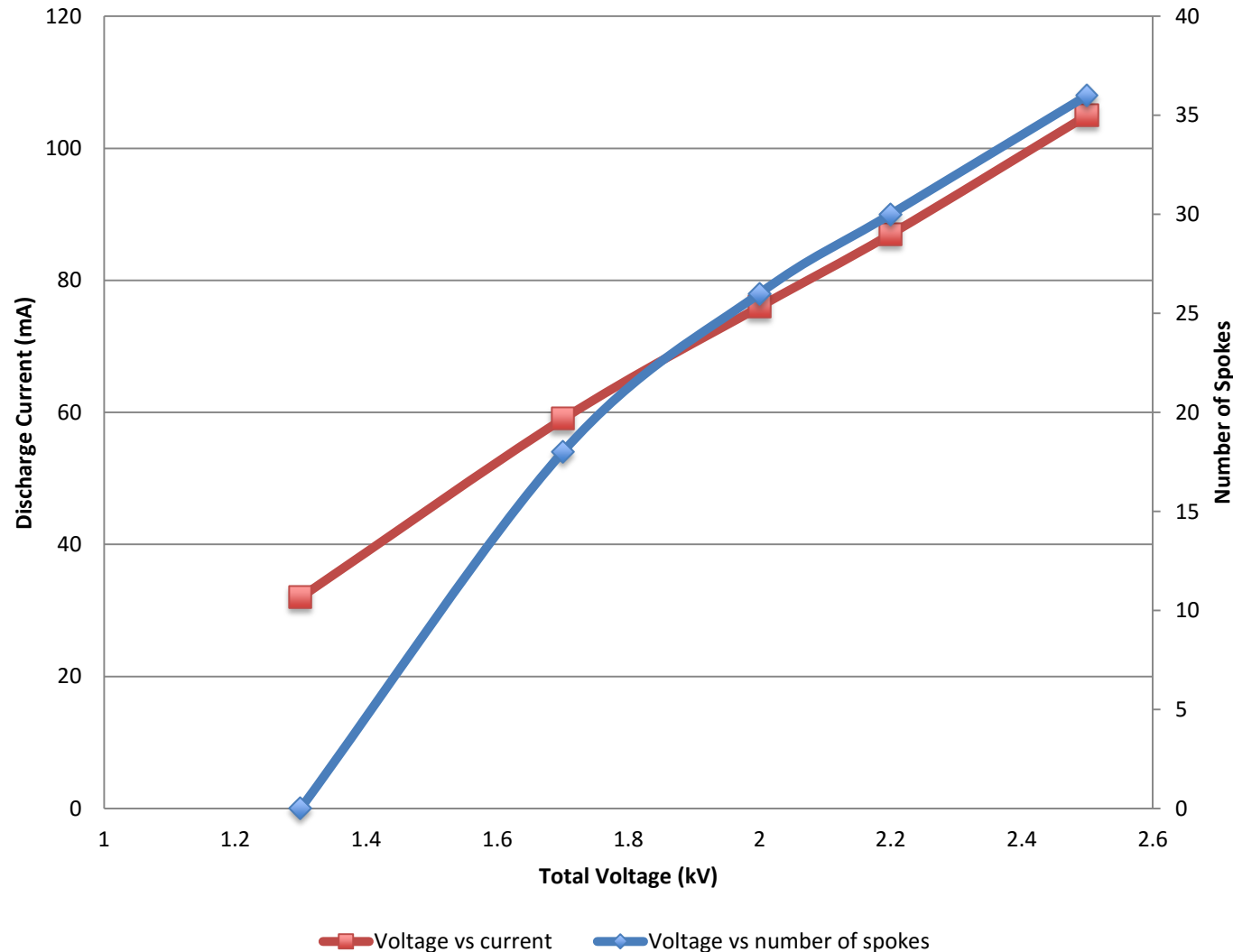
Sodium Halo



- Self-organization patterns starts with ring pattern at lower power and evolves to spoke pattern at higher power (on the surface of NaCl solution).
- Yellow halo observed as Na vapor is introduced into the column at higher powers
  - Suggests localized evaporation
  - Na may play role in modifying discharge column impedance (feed back) and perhaps plays a role in pattern formation



# NaCl solution II: IV curve and Spokes number variation

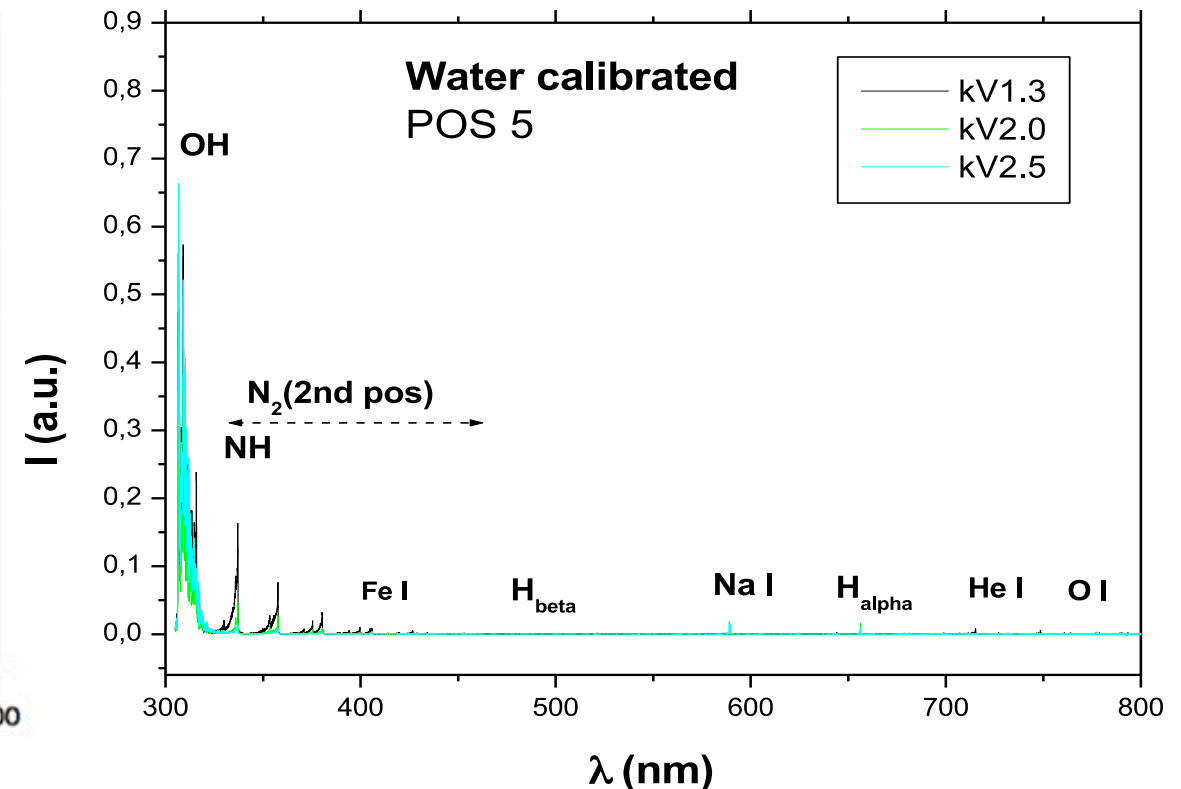
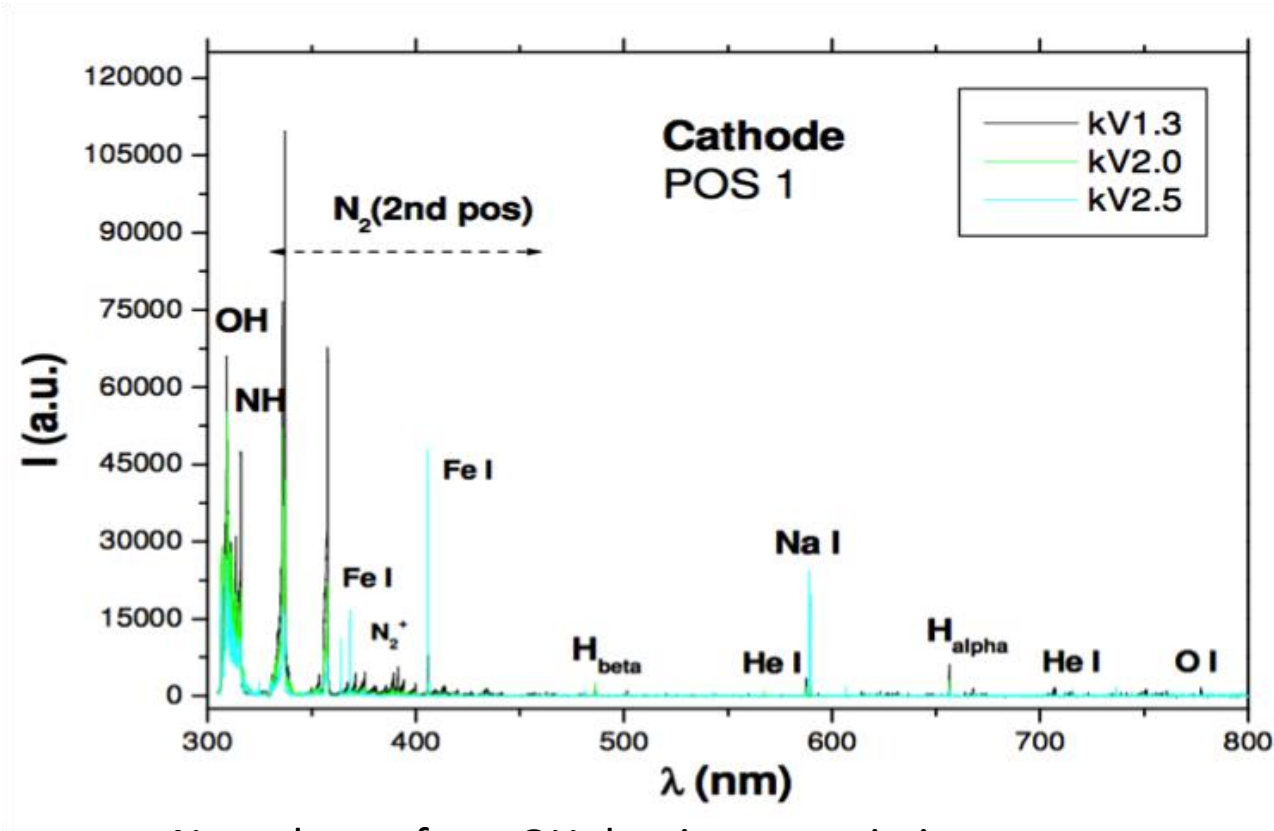


- Above 1.3 kV, the number of spokes increasing linearly with total voltage.
- The spoke number is also linear with increasing discharge current.
- Presumably some threshold current is required before spokes can actually form.
- Data suggests that pattern is necessary for current continuity—to support more current—more spokes

# NaCl III: Optical Emission Spectra(OES)

## ◆ Spatially resolved optical emission spectroscopy

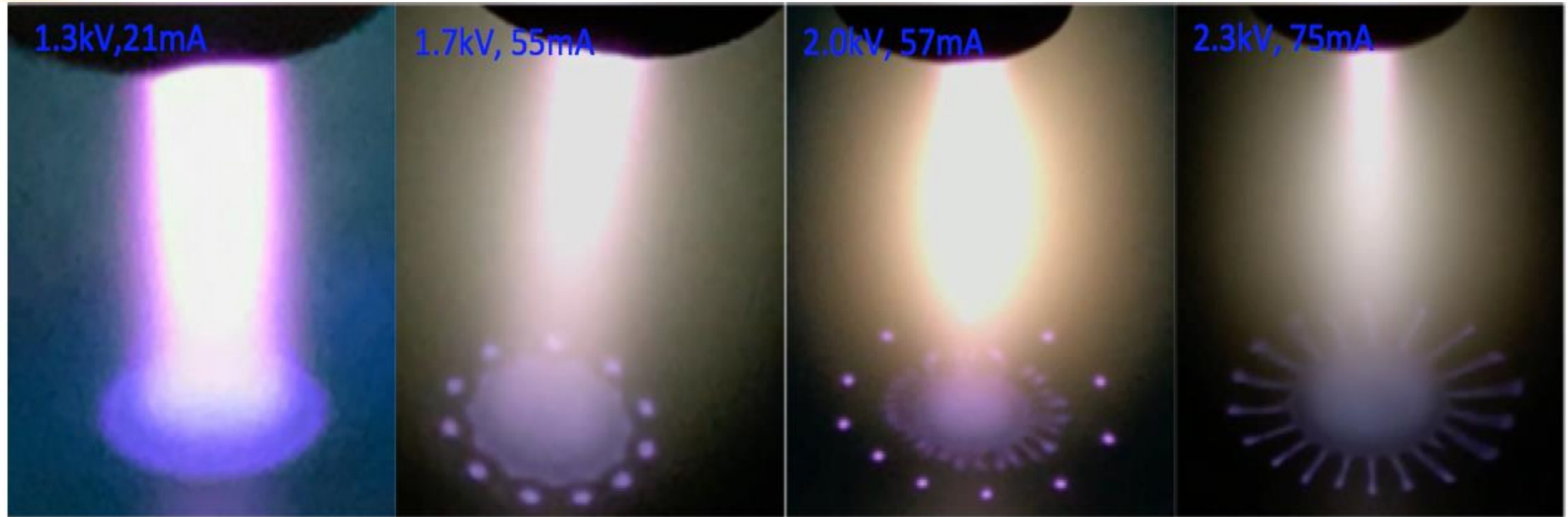
- Position 1 is in gap length near to the cathode 8 mm above the surface.
- Position 5 is near to the liquid surface.



- Near the surface, OH dominates emission
- Intense emission near cathode due to energetic electrons

# CuSO<sub>4</sub> I:

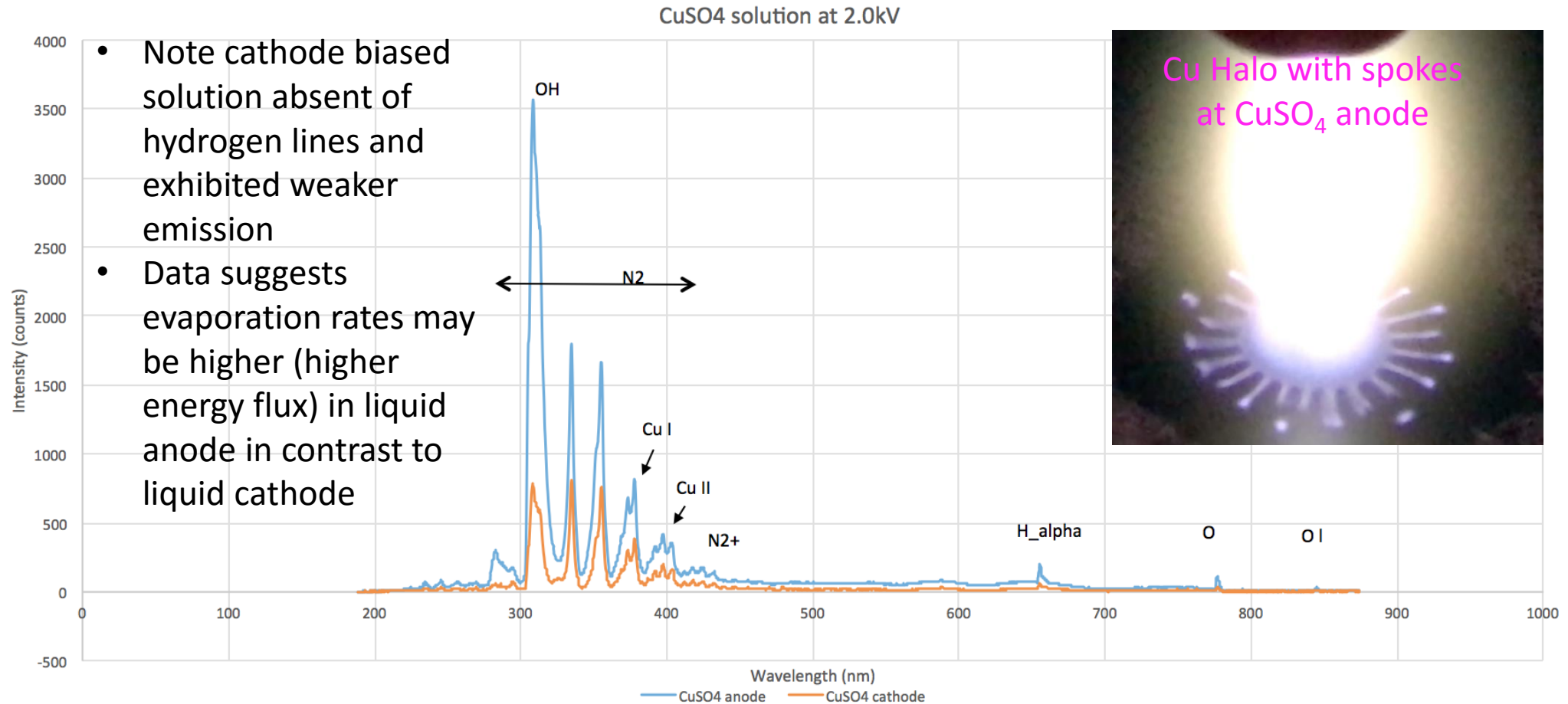
## Self-organization with He Plasma Injection



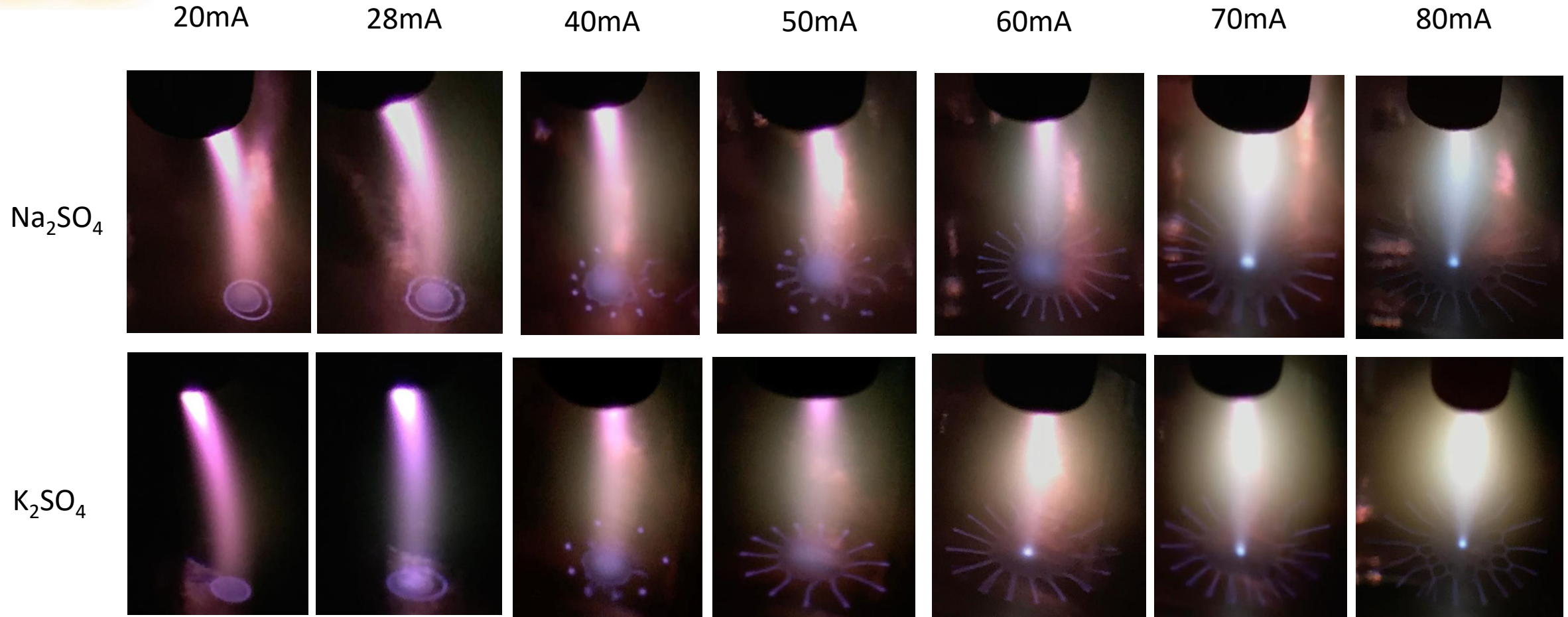
- ◆ More complex shaped self-organization patterns observed on the the surface of CuSO<sub>4</sub> solution:
  - Lower applied voltage shows the dots around of main plasma.
  - At 2.0 kV but lower current (57mA), the pattern shapes appeared as a transformation between the dots and spokes .
  - Similar to NaCl solution, no patterns were found in lower voltage(1.3 kV) besides the plasma attachment, and the same spokes were observed in higher applied voltage.

# CuSO<sub>4</sub> II: OES with Different Liquid Polarity

- ◆ Spectrum of plasma glow near to the CuSO<sub>4</sub> surface acquired with different polarity (cathode or anode) were measured at 2.0 kV with Cu halo emission.



# PSTL Self-organization Patterns in Constant Current Mode I



- ◆ For the salt solution associated with  $\text{SO}_4^{2-}$ , the self-organization patterns were acted in the same behavior which from double ring in lower current to dots at 40mA, then to large spokes in higher current region.

# Self-organization Patterns in Constant Current Mode II

20mA

28mA

40mA

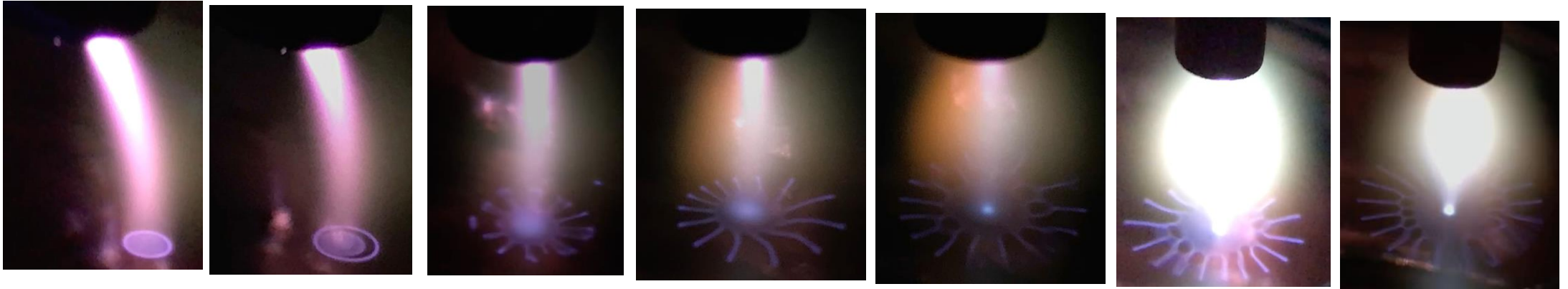
50mA

60mA

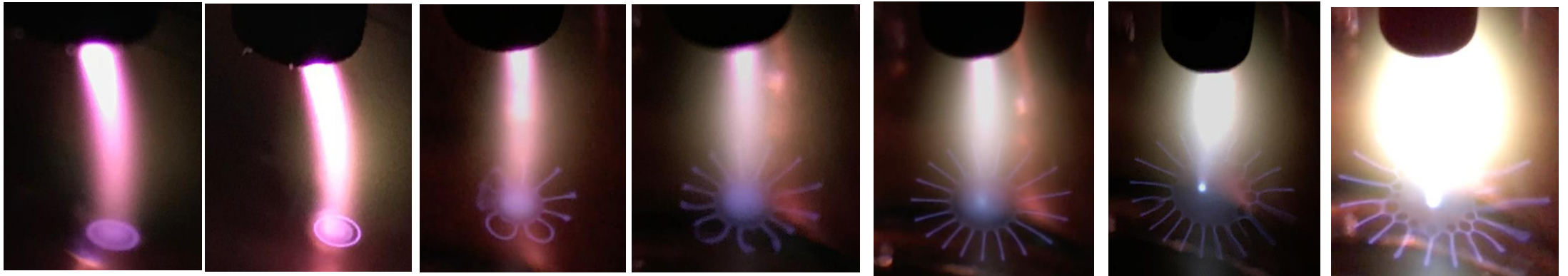
70mA

80mA

NaCl

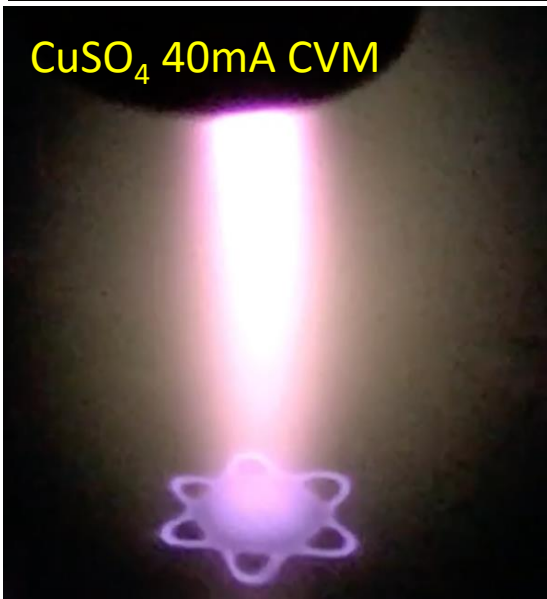


KCl



- ◆ For the salt solution associated with  $\text{Cl}^-$ , the self-organization patterns were acted in the same behavior which from double ring in lower current to large spokes in higher current region.

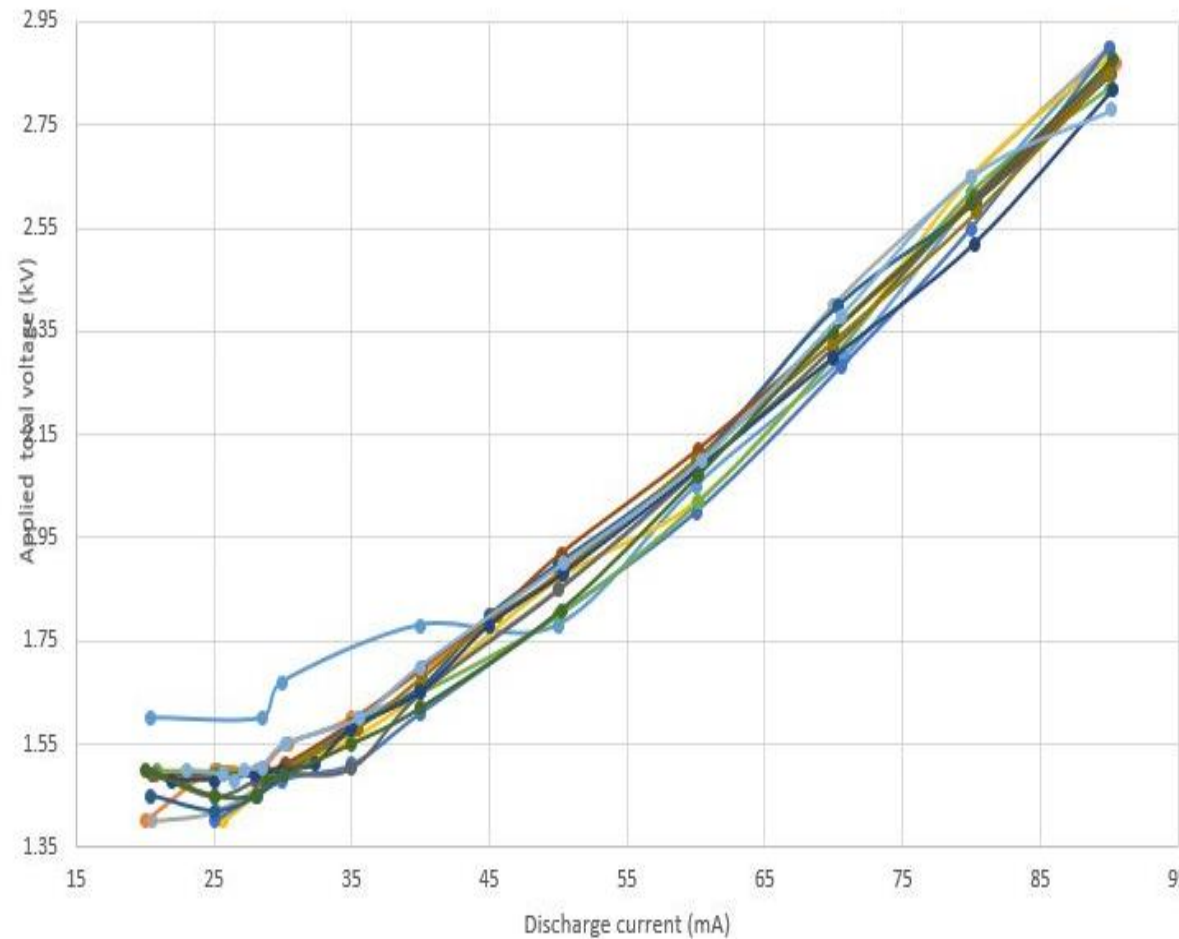
# New Self-organization Patterns at Lower Current Region



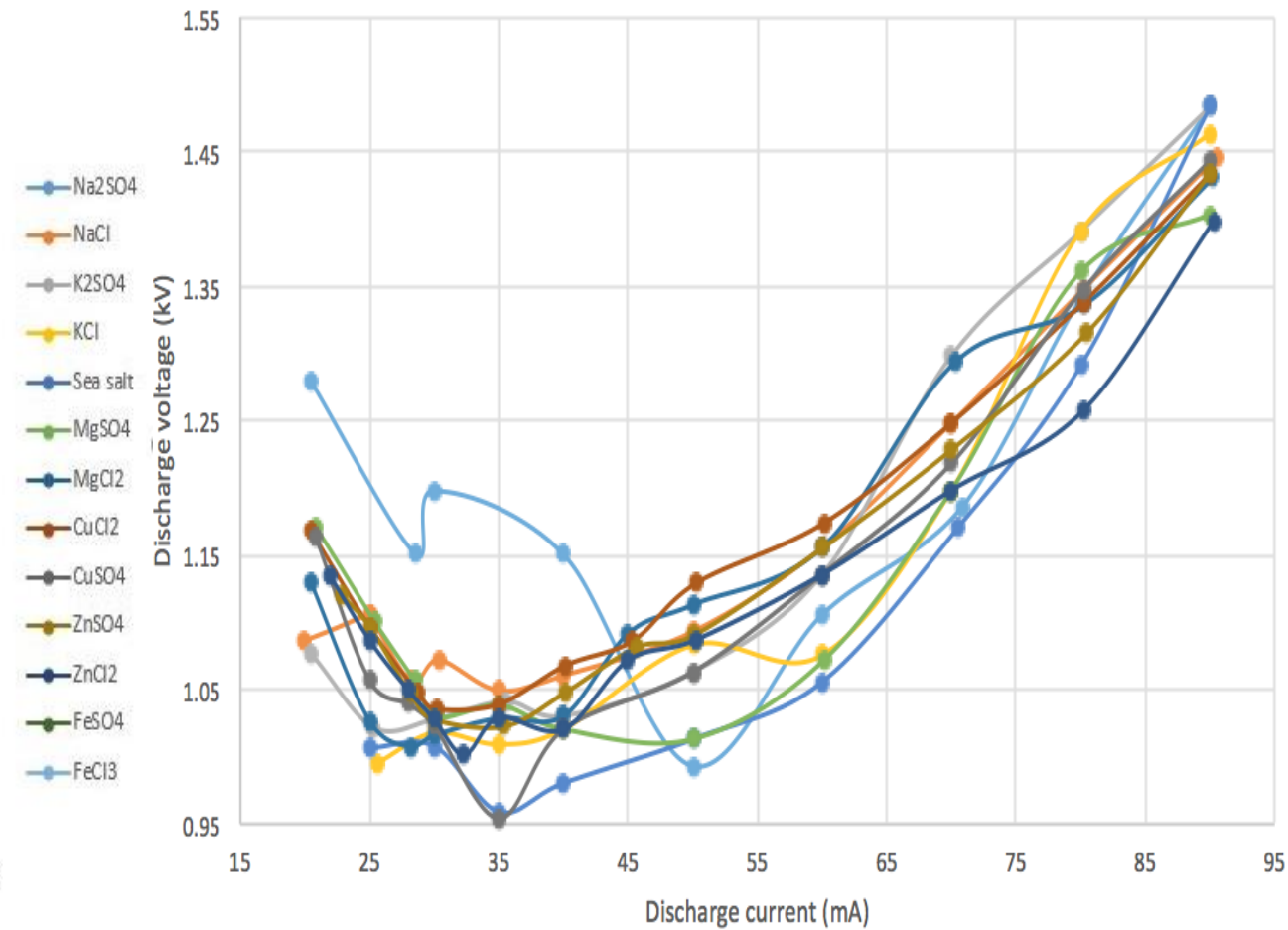
- ◆ At lower current region, more complex shapes were observed in different salt solutions.
  - ✧ In constant current mode, flower shaped self-organization structures were observed around of 30 mA. A network structure observed during reverse discharge (high to low).
  - ✧ In constant voltage mode, a star shaped self-organization structure were found on the surface of copper sulfate solution.
  - ✧ These results suggest that the ion mobility may play a role in self-organization formation.

# Discharge Characteristics of Each Salt Solution

Total Voltage Variation



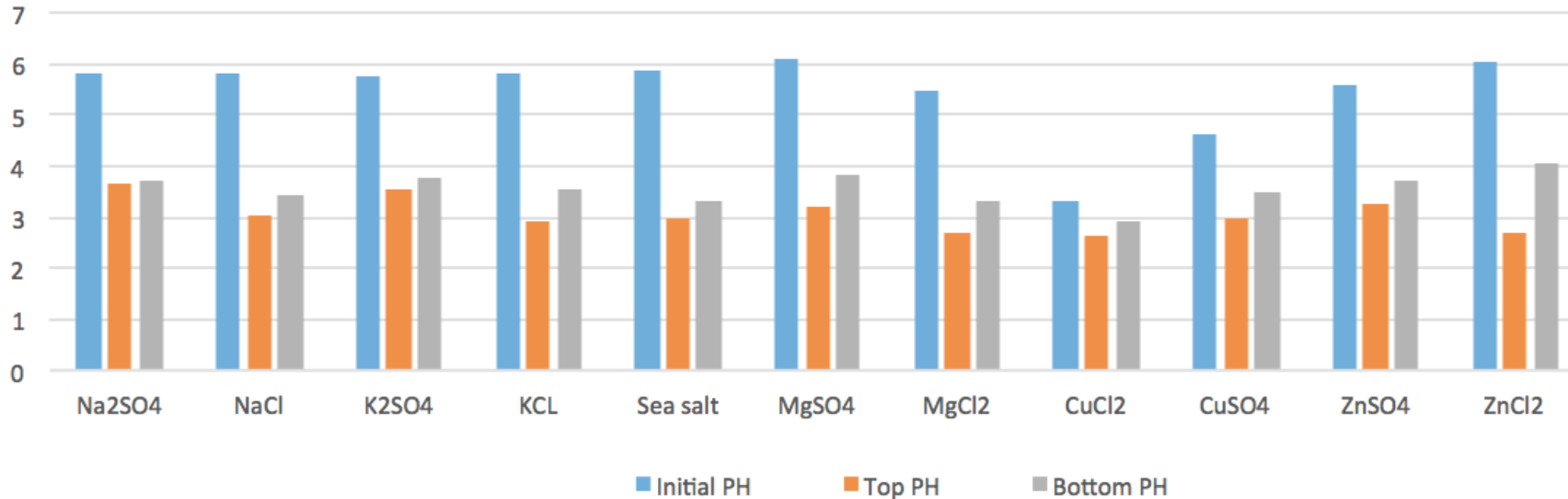
Discharge potential variation



Na<sub>2</sub>SO<sub>4</sub>  
 NaCl  
 K<sub>2</sub>SO<sub>4</sub>  
 KCl  
 Sea salt  
 MgSO<sub>4</sub>  
 MgCl<sub>2</sub>  
 CuCl<sub>2</sub>  
 CuSO<sub>4</sub>  
 ZnSO<sub>4</sub>  
 ZnCl<sub>2</sub>  
 FeSO<sub>4</sub>  
 FeCl<sub>3</sub>

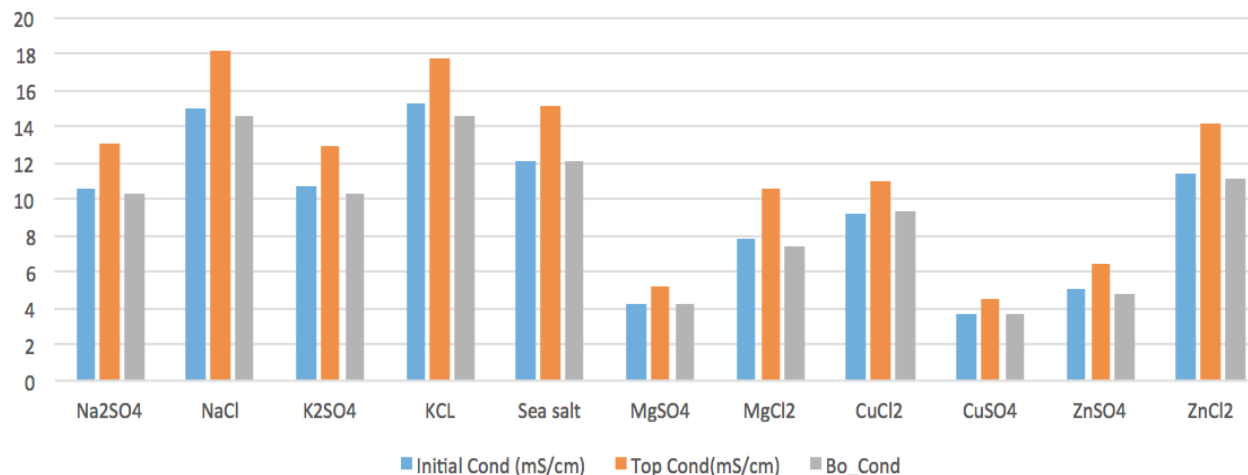
# Chemistry properties from different electrolytes

PH values

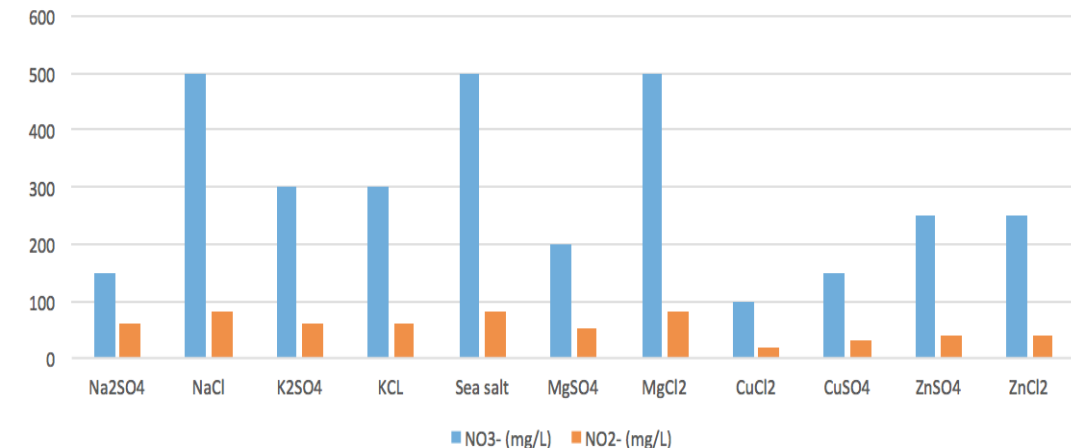


- ◆ All solution PH reduced after 20 minutes He plasma injection. However, the bottom PH values are higher than the PH on the surface of solution.
- ◆ All solution conductivities went up after the test but opposite with PH measurements.

Conductivity (mS/cm)



Nitrate, Nitrite





# Relationship Between Low Pressure and High Pressure Anode Spots

- At low pressure, anode spots appear on electrodes under certain conditions
  - If electron loss rates greatly exceed ion losses near electrode a double layer will form
  - If field is high enough ionization can take place in the sheath and anode spots forms and expands into plasma
- In 1 atm glows, electron diffusion from cathode region to anode is tortuous.
  - Electrons may need to be produced locally owing to large surface electric fields
    - This would also leave positive space charge region adjacent to sheath region thus perhaps a double layer structure exists near surface as well
    - How does presence of evaporated metal ions affect this structure?
    - Is space charge repulsion the reason for spoke like structures associated with positive space charge?
  - Spots inherently increase the collection surface area for electrons; double layer virtual cathode can trap electrons thus increase electron density locally—and thus may be necessary to maintain the discharge

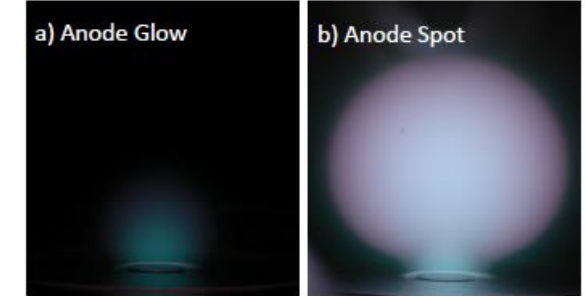


FIG. 2. Photograph of (a) an anode glow and (b) an anode spot.

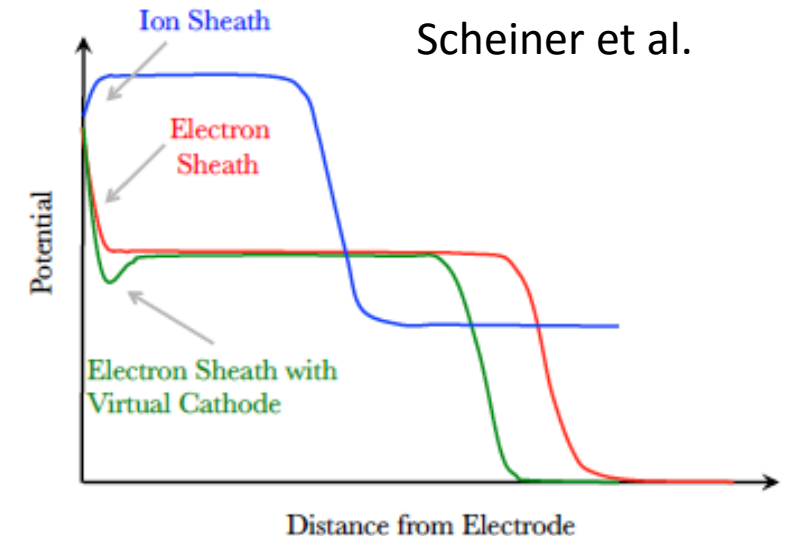


FIG. 12. Sketch of three types of potential structures associated with an anode spot.



# Future Work and Acknowledgements

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## Future work

- High resolution spatially resolved emission will be carried out for both copper sulfate and sodium solutions
  - Need to elucidate nature of self absorption in sodium and copper vapor clouds
  - What role does the metal vapor play in discharge maintenance and cloud formation?
- Estimate fall voltages at anode surface
  - Yield insight into evaporation and liquid phase chemistry
- What is the plasma density in spokes and spots?

## Reference

- J.P. Trelles, J.Phys. D: Appl. Phys. 49 (2016) 393002.
- Staack, David, Bakhtier Farouk, Alexander Gutsol, and Alexander Fridman. "Characterization of a dc atmospheric pressure normal glow discharge." *PLASMA SOURCES SCIENCE AND TECHNOLOGY* 14 (2005) 700-711.

## Acknowledgements

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