HONEYCOMB DISCHARGES – THEIR PHYSICAL PROPERTIES AND CHEMICAL POTENTIAL FOR ENVIRONMENTAL APPLICATIONS

Karol Hensel[†], **Gokul Selvaraj, Richard Cimerman, Mario Janda**

Faculty of Mathematics, Physics and Informatics, Comenius University, Bratislava, Slovakia

[†]Presenter: Karol Hensel
[*]Corresponding author's e-mail: *hensel@fmph.uniba.sk*

ABSTRACT

Plasma catalysis provides a beneficial combination of nonthermal plasma (NTP) with catalysis that has many potential environmental applications including air pollution control. Plasma catalytic reactors usually utilize dielectric barrier discharges packed with catalyst pellets, where discharge propagates in a contact with the pellet catalyst and maintains catalytic surface reactions. Another option for plasma catalytic reactor represents combination of NTP with honeycomb catalysts. From the perspective of effective surfaces involving chemical reactions, pellets have a structural disadvantage over honeycombs, as they provide high surface-to-volume ratio, low pressure drop and high mass and heat transfer. Generation of stable discharges in channels of the honeycomb represents a major challenge in honeycomb discharge from a practical perspective. In last two decades, honeycomb discharges have been studied both experimentally and theoretically [1]. In many studies glass capillary tubes were used for discharge visualization and emission spectroscopy. Numerical simulations helped to understand discharge generation, and propagation dynamics inside honeycomb monoliths and capillary tubes, and reported in the effects of tube radius, dielectric constant, gas humidity, etc. The chemical activity of the discharge was tested for O_3 production, hydrocarbons removal and the regeneration of diesel particulates [2, 3].

This contribution summarizes our results on generation of discharges in capillary tubes and honeycomb monoliths. Electrical and optical measurements were performed in various geometry (diameter, length, cpi), gases (air, N2, O2, H2O) and power supplies (AC, DC, pulsed). The results showed dominant effect of tube diameter and humidity on discharge propagation velocity [4]. Tests with a bundle of capillary tubes were performed to assess the overall stability and spatial homogeneity of the discharge. The discharge was generated by a combination of DC an AC discharges in a pellet bed [4] or in a multi-hollow DBD [5]. The homogeneity and the stability largely depended on the discharge polarity, ballasting resistor, and gas humidity. At last, the discharges generated in honeycombs monoliths of various geometry (length, cpi) were tested. They were also briefly subjected to the investigations of its plasma chemical activity (generation of O_3 and toluene removal).

Keywords: honeycomb discharge, capillary tubes, plasma catalysis, exhaust gas cleaning

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Karol HENSEL, Gokul SELVARAJ, Richard CIMERMAN, Mário JANDA

Faculty of Mathematics, Physics and Informatics, Comenius University, Bratislava, Slovakia
hensel@fmph.uniba.sk

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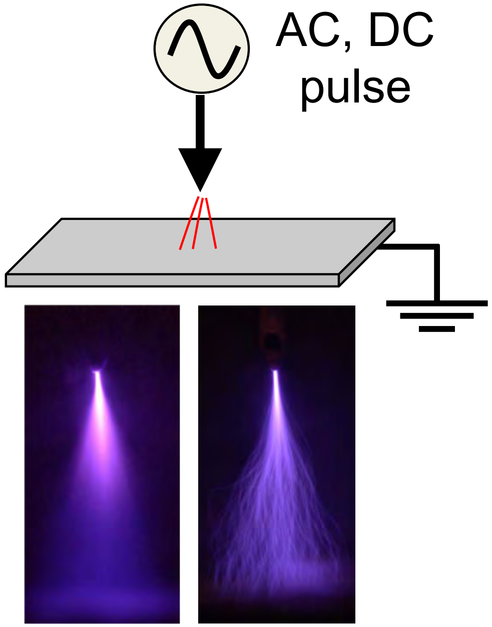
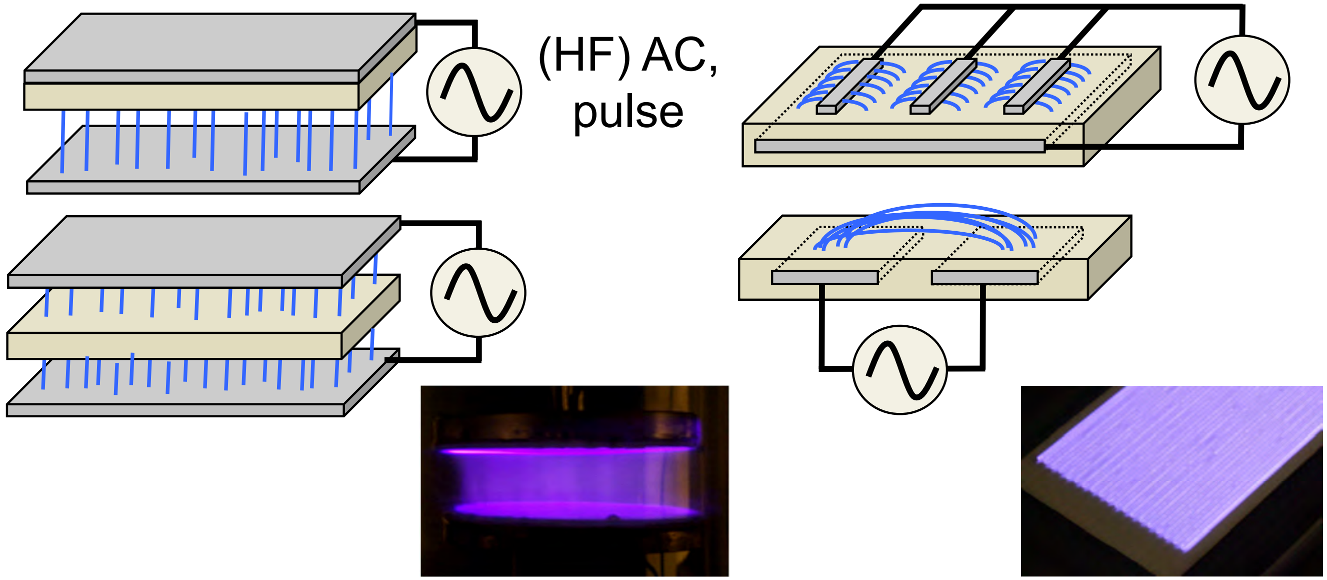
Atmospheric discharges

dielectric
electrode

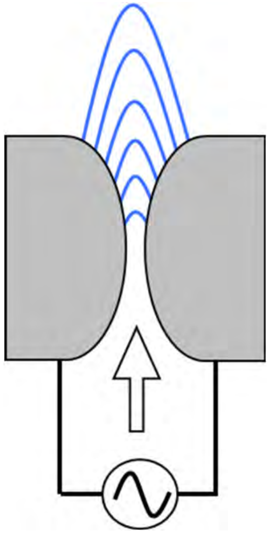
corona discharge



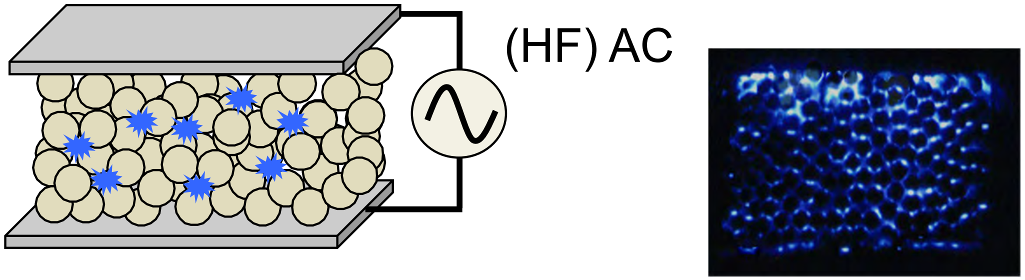
barrier volume/surface discharge



gliding arc discharge



discharge in pellet bed



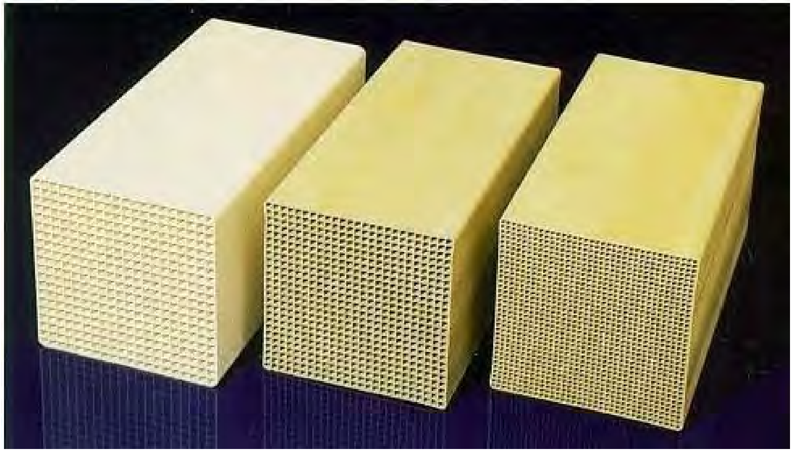
Shapes of materials/dielectrics/catalysts

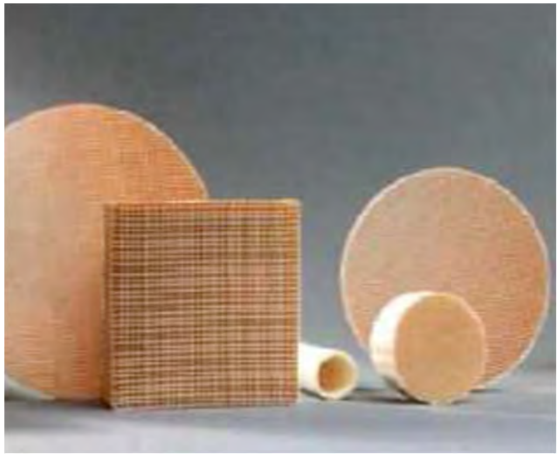
pellets
(1 – 5 mm)

foams
(1 – 100 μm)

foams
(100 μm – 1 mm)

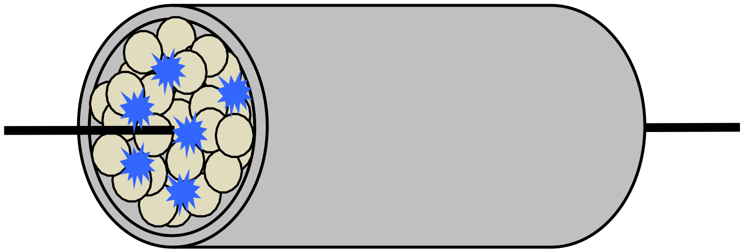
tubes
(10 μm – 1 mm)



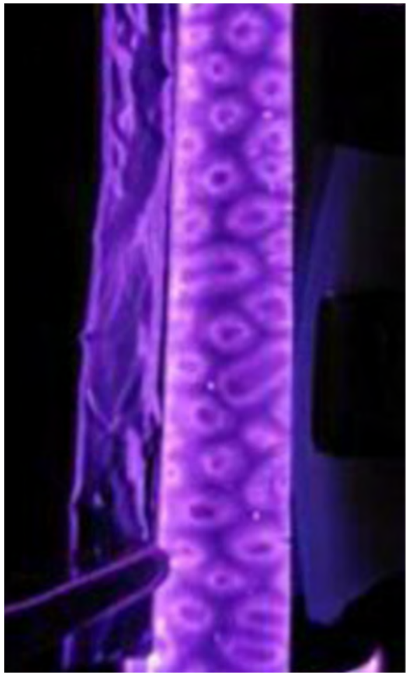


honeycomb monoliths
(100 μm – 1 mm)

Interaction of discharge with material/catalyst



discharge plasma vs. material/catalyst



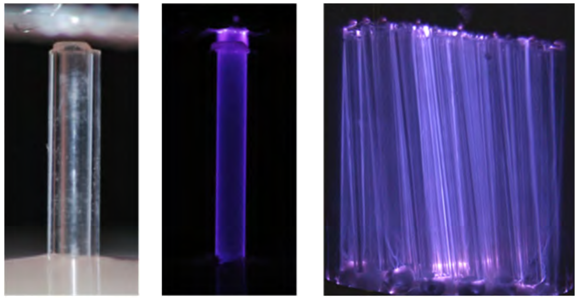
- Their interaction affects the chemical process (removal efficiency, selectivity, carbon balance).
- **Catalyst effect on discharge** → change of electric field; surface discharge distribution; adsorption of pollutants; …
- **Discharge effect on catalyst** → UV activation; lattice oxygen activation; catalyst dispersion; plasma induced desorption; …

- How a material (structure, dimensions, catalytic properties) affects the discharge ignition, propagation and chemical activity?

Overview of presented results

- Summary of our results on generation of discharges in glass capillary tubes, ceramic honeycomb monolith, ~~ceramic foams.~~

discharge in capillary tubes



discharge in honeycomb monoliths

- All tested systems were of point-to-plane reactor geometry.
- **Materials** of various geometry (diameter, length, cpsi), various **power supplies** (AC, DC, pulsed) in both polarities, various **gases** (air, N_2, O_2).
- **Electrical diagnostics** included I/V measurements and power evaluation.
- **Optical diagnostics** included photography, emission spectroscopy as well as photomultiplier and fast camera imaging.
- **Chemical activity** was briefly investigated by absorption spectroscopy.

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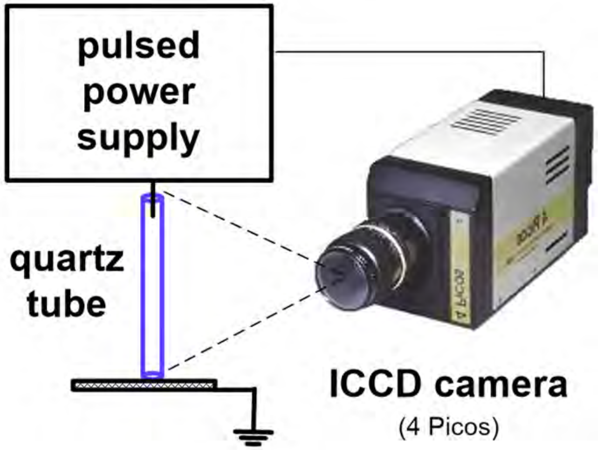
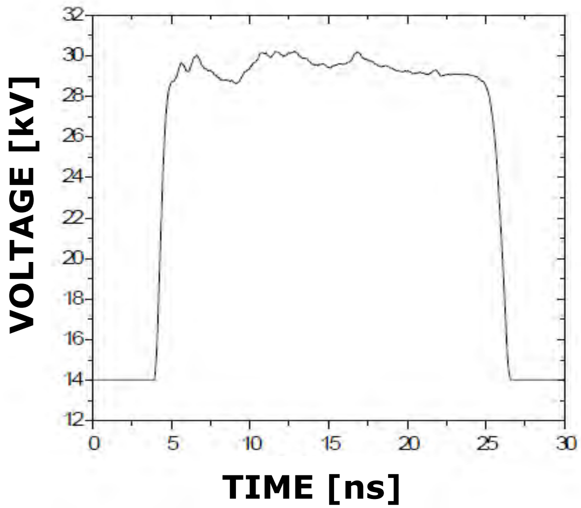
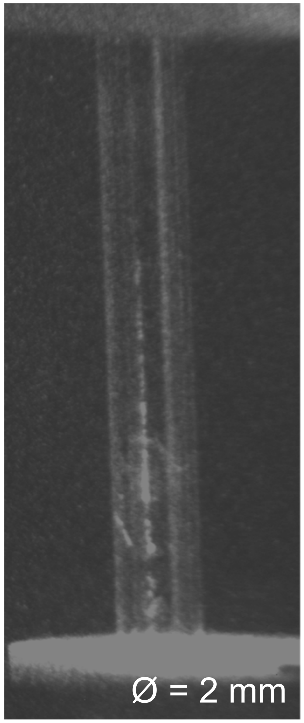
K. Hensel, et al. IEEE Trans. Plasma Sci. 36, 1282 (2008)

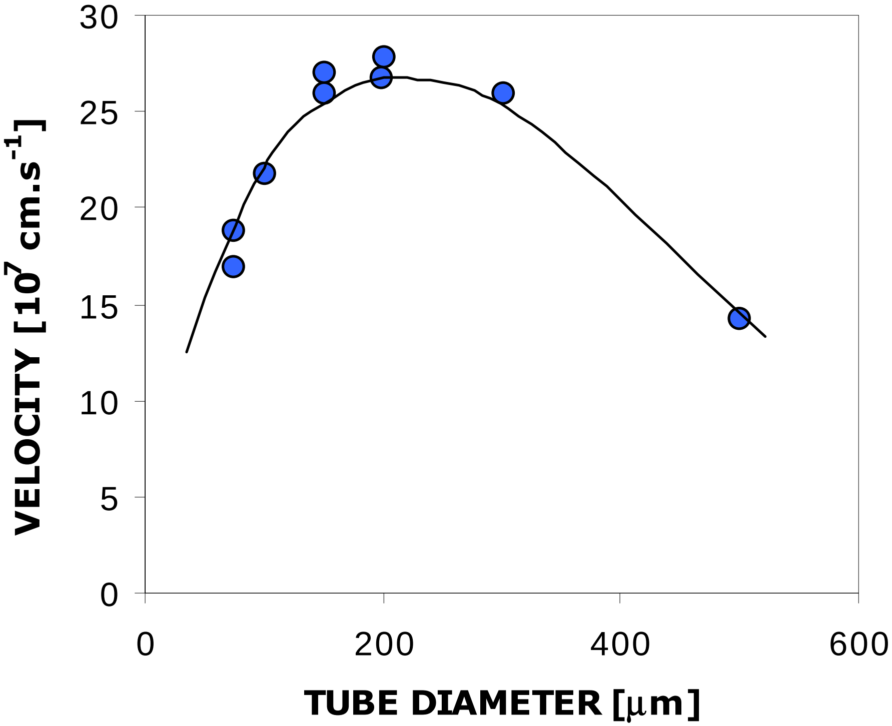
Single tube – Pulsed HV





- Tube diameter affects both the structure and the velocity of the discharge.
- Structure: tubular → homogeneous.



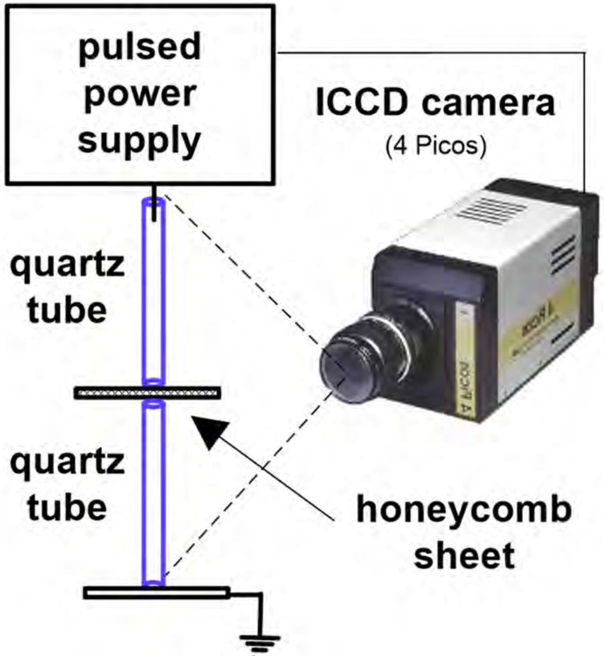


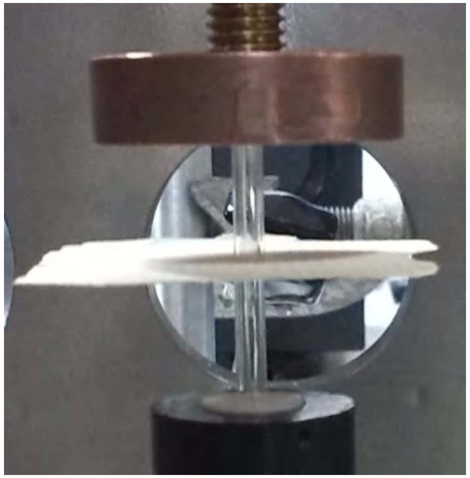
Tubes → dimensions: Ø 50 μm – 2 mm
Electrode → dimensions (material): Ø 50 μm
Pulse → amplitude 7-30 kV, duration 20 ns
Gas → ambient air

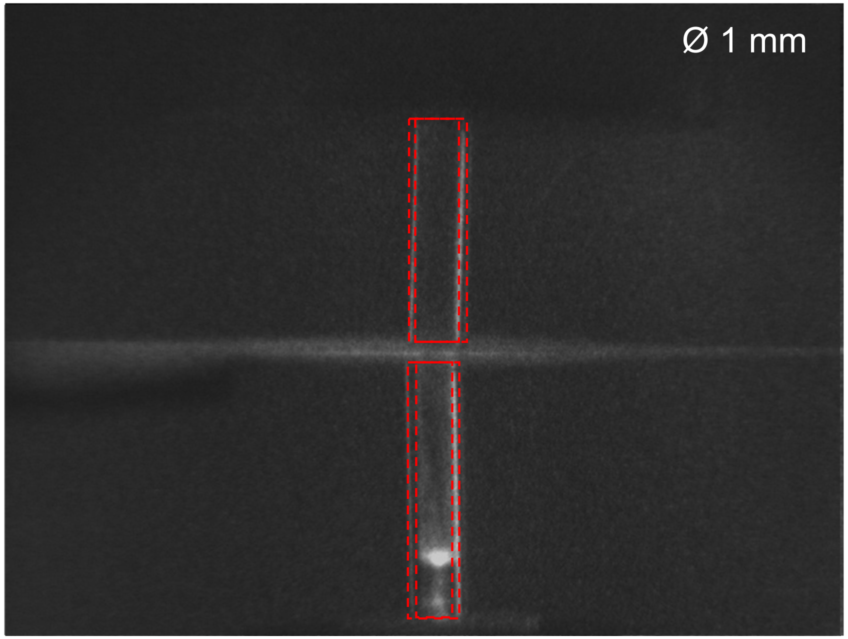
Discharge propagation velocity *vs.* tube diameter

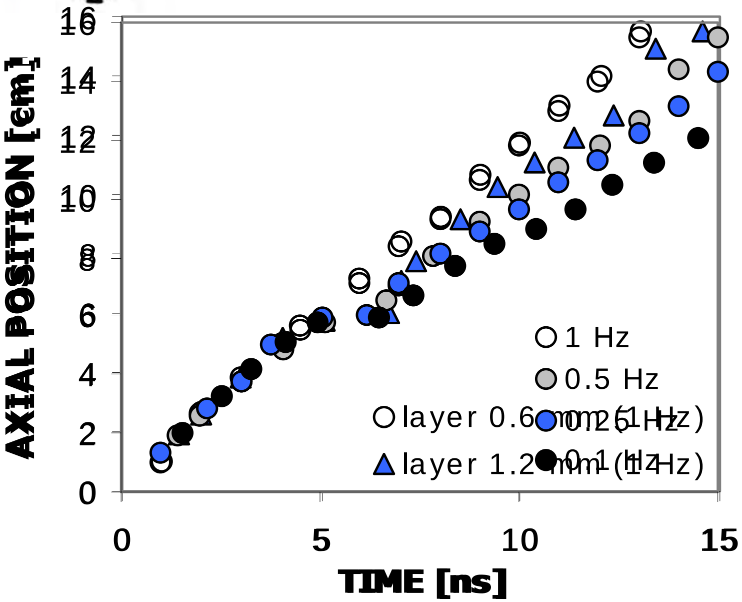
- Propagation velocity linearly ↑ with applied voltage ↑ and with ε_r ↓ .

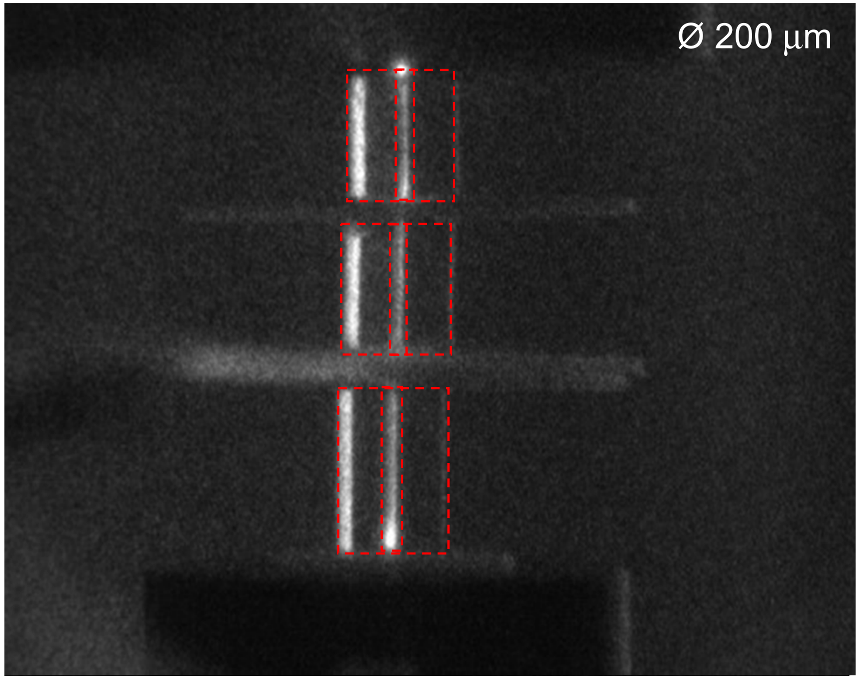
Single tube + honeycomb sheet – Pulsed HV





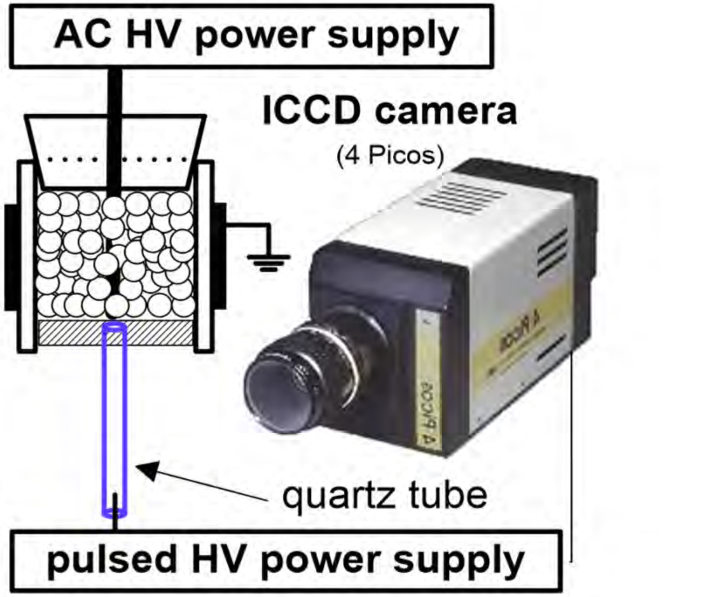


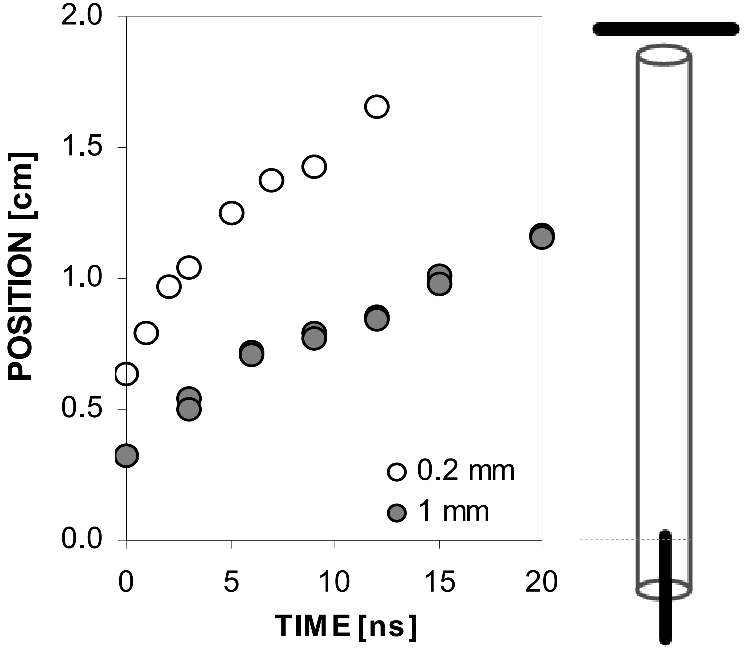


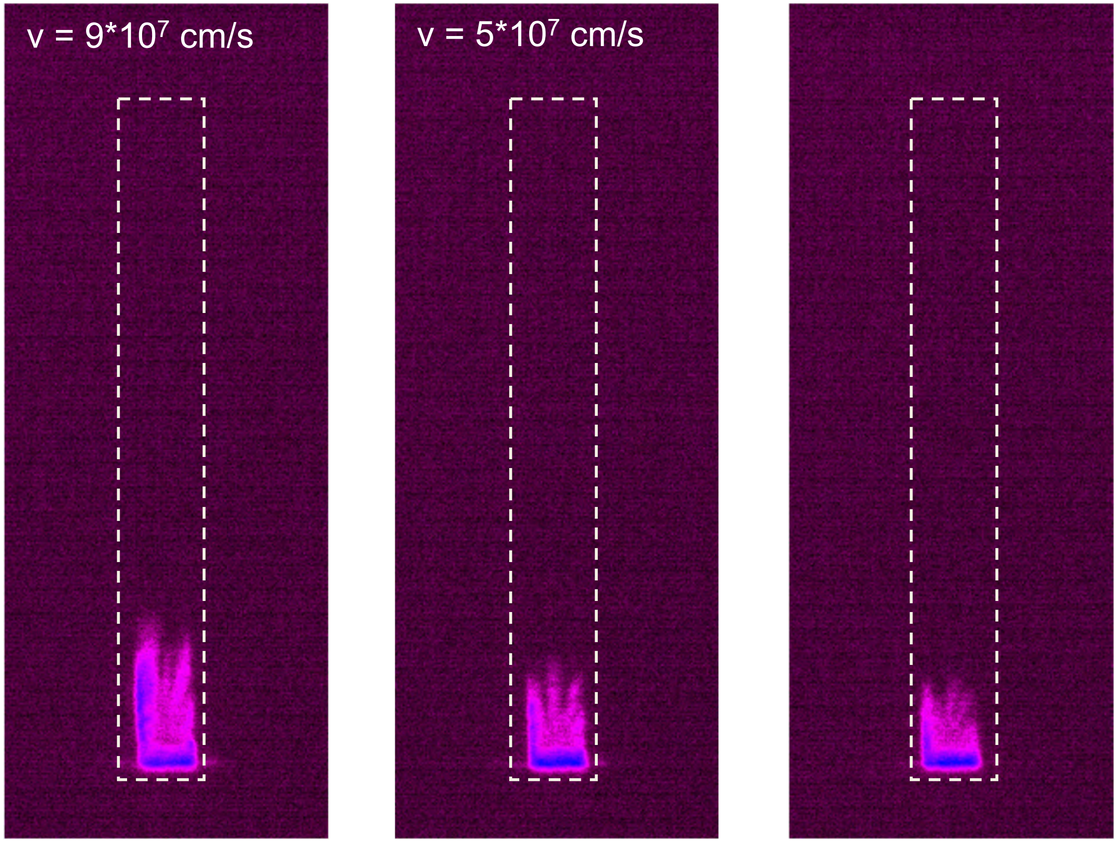


Discharge propagation velocity

Single tube – Pulsed HV + AC HV





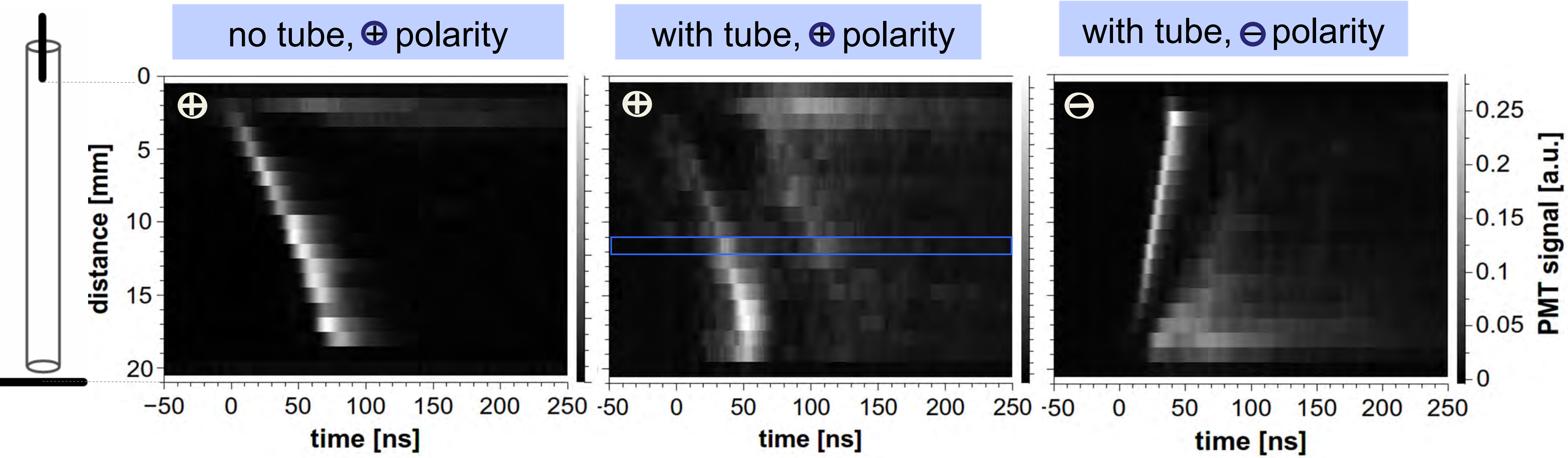


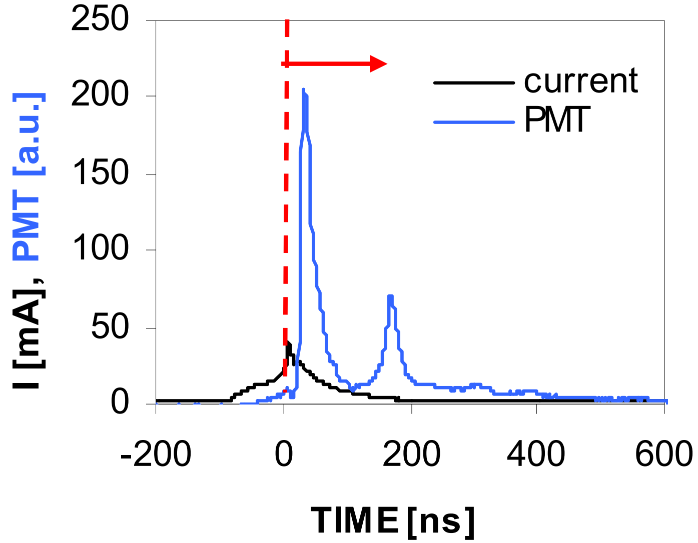
U_{AC}= -13 kV U_{AC}= 0 kV U_{AC}= +13 kV

Discharge propagation toward cathode
vs. time *vs.* AC applied voltage
[∅ = 1 mm, L = 3 cm, U_{pulse} = +15 kV, Δt_{pulse} = 20 ns]

Single tube – AC HV

- Spatio-temporal evolution of discharges by a photomultiplier tube (PMT) on a translation stage enabled mapping the light emission along the axis of the tube.

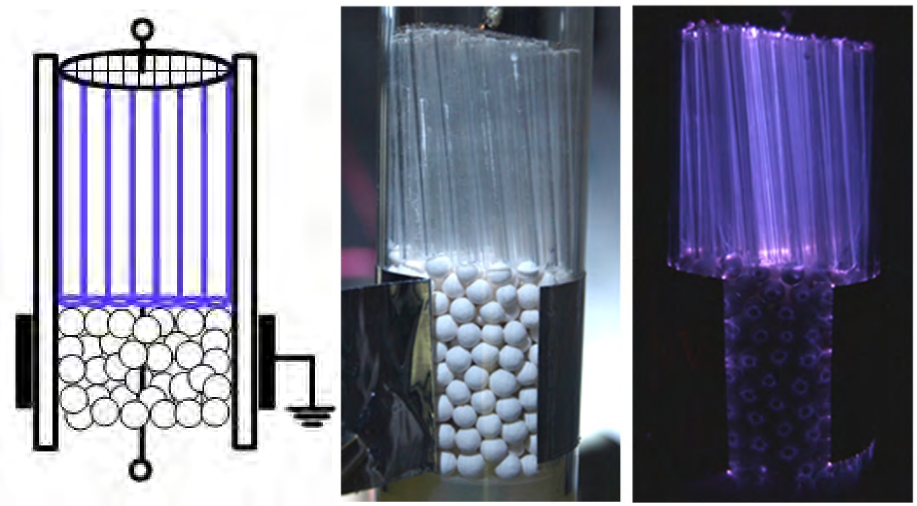


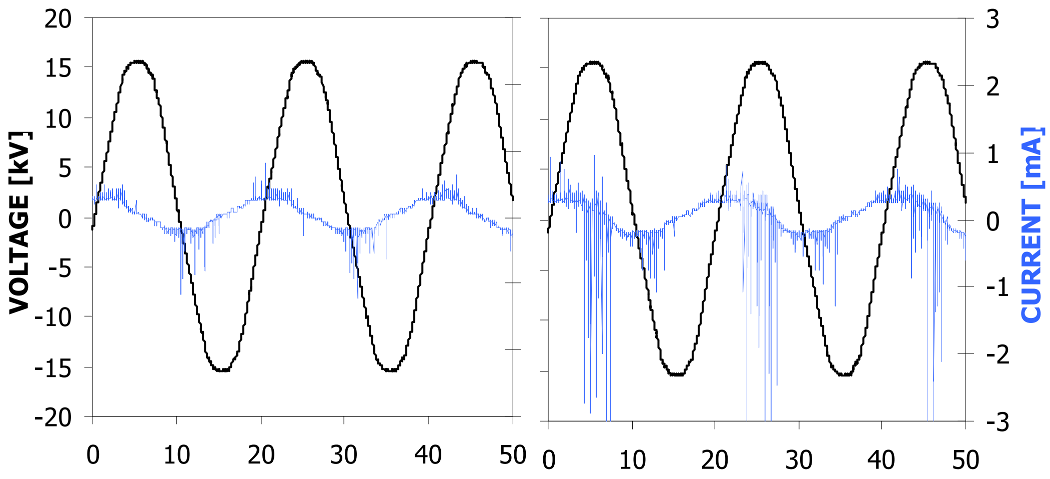


Discharge propagation with and without tube
[L = 1.8 cm, Ø = 2 mm, U_{AC} = 13 kV]

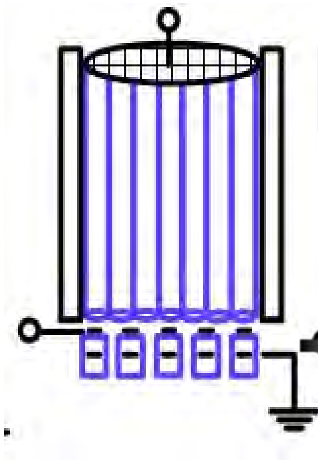
- Several light trails in one sequence.
- Propagation was faster in negative polarity.
- Peak currents ↑ tube diameter (Ø) ↓.
- Discharge propagation velocity ↑ tube diameter (Ø) ↓.

Bundle of tubes – DC and AC HV

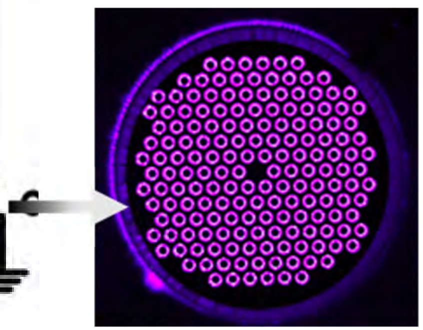


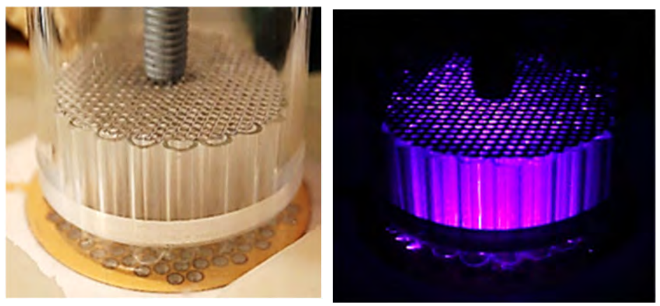


Discharge assisted by AC **pellet bed discharge**



multi-hollow DBD

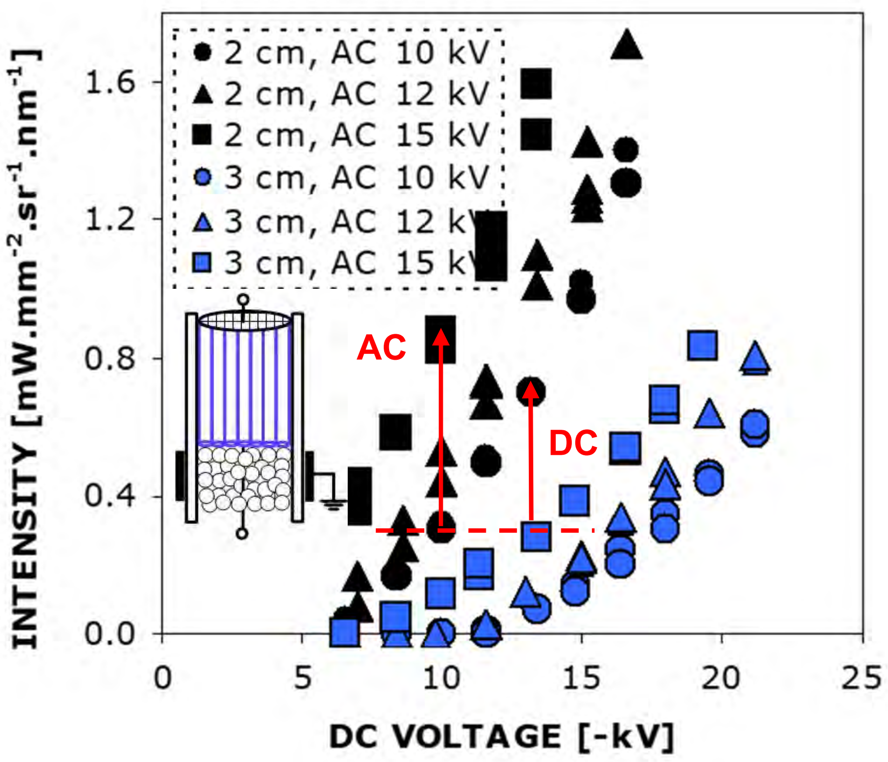




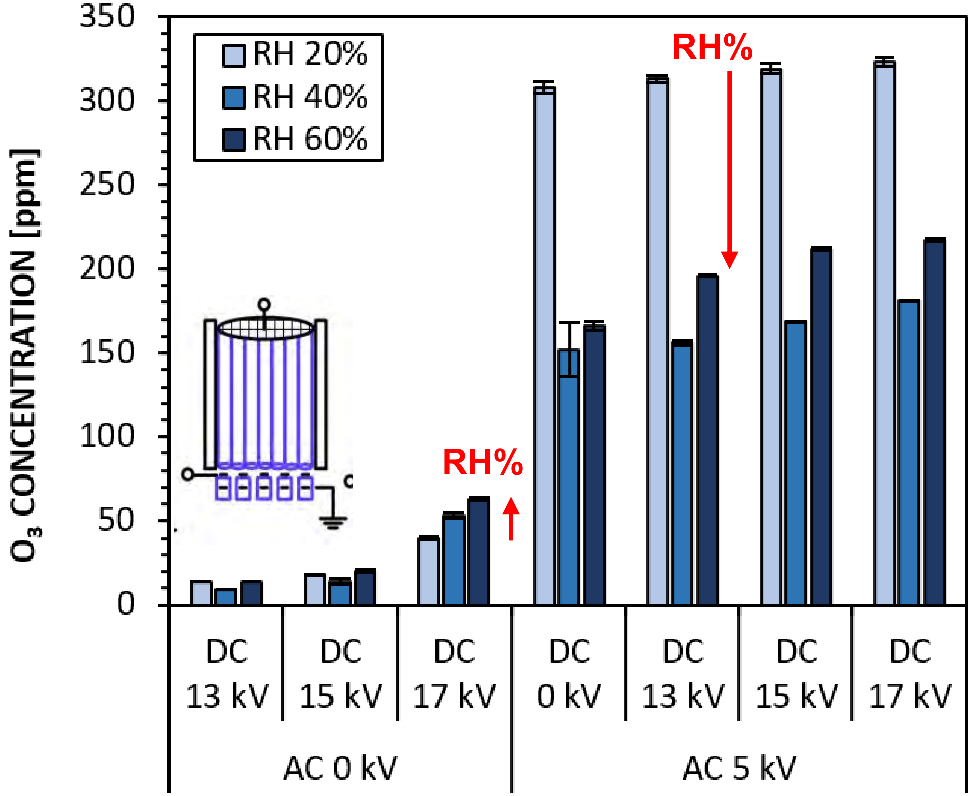
Discharge assisted by AC **multi-hollow DBD discharge**

- The auxiliary AC discharges served as ionizers, while DC bias (U_{DC}) extended the discharges inside the capillary tubes.

Bundle of tubes



Emission intensity *vs.* DC and AC voltages *vs.* tube length (L) [**pellet bed**, Ø 2 mm]



O_3 production *vs.* DC and AC voltages *vs.* RH% [**micro-hollow SDBD**, Ø 2 mm]

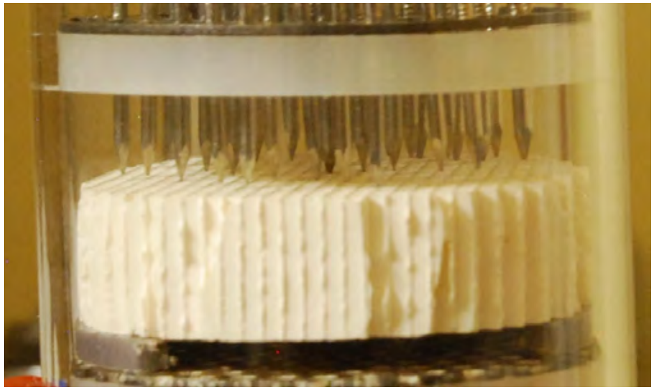
- The homogeneity, stability as chemical activity of the discharges strongly depended on the applied voltages (U_{AC}, U_{DC}), and feed gas humidity (RH%).

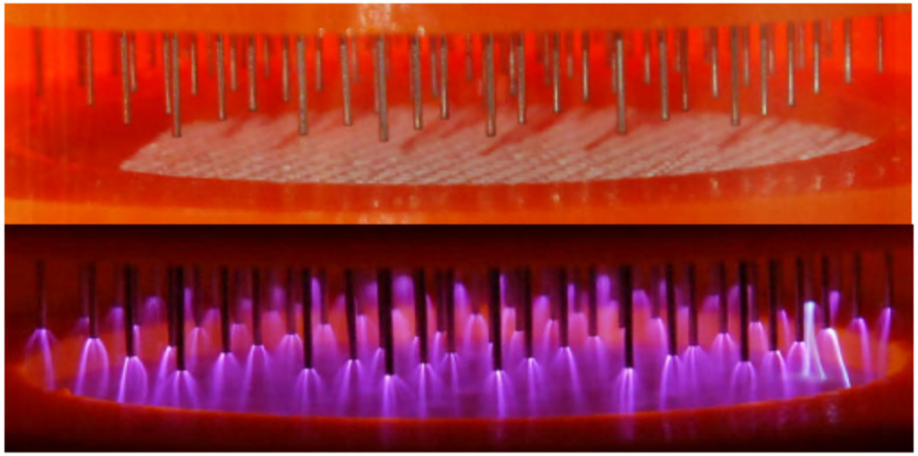
Honeycomb monolith

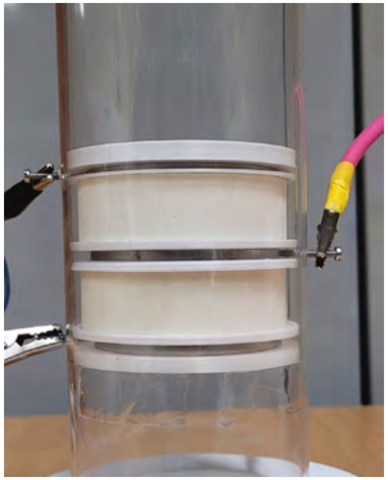
- Honeycomb discharges in various geometries [L = 12 mm, CPSI = 64/200, d = 1-5 mm] was tested for its properties and chemical activity.

CPSI = cells per squared inch
d = distance from HV electrode to honeycomb surface

Discharge in honeycomb monolith
[d = 5 mm, L = 17 mm, U_{DC} = 21 kV, CPSI = 200]

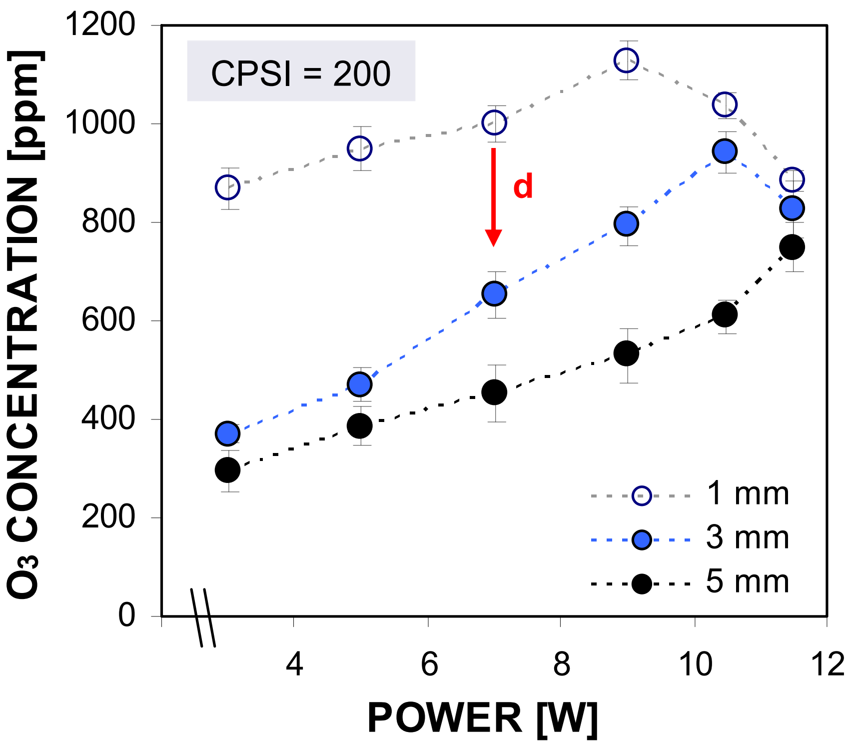


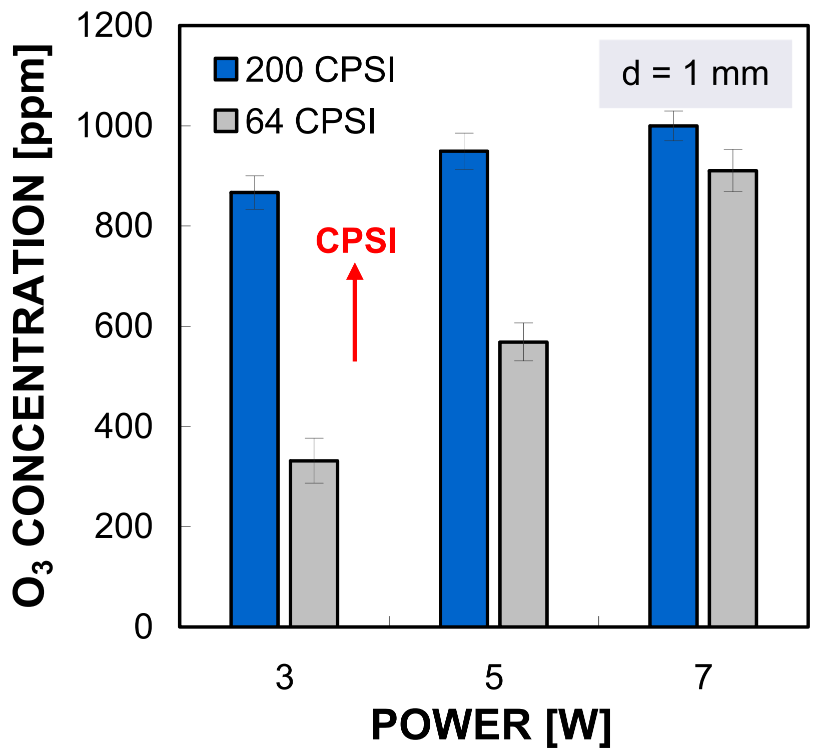




Discharge in honeycomb monolith
[d = 0 mm, L = 15 mm, U_{DC} = 18 kV, CPSI = 200]

Honeycomb monolith





O_3 production *vs.* discharge power (P) *vs.*
pin-to-honeycomb gap (d) vs. CPSI [air, 0.25 L/min]

- Ozone production ↑ discharge power (P) ↑ and CPSI ↑, and with pin-to-honeycomb gap (d) ↓ and gas flow rate ↓ (Q).

Summary

- Atmospheric **discharges in contact with materials** (dielectric/catalytic) are important for many plasma applications
(air pollution control, fuel reforming, surface treatment and modifications).
- Understanding **plasma–catalyst interaction** is important for process optimization.
- We investigated discharges in a **single** capillary tube, bundle of tubes and honeycomb monolith generated by various power supplies and their combination.
- Effects of **tube diameter** (Ø), **discharge gap** (L), **type of voltage** (U_{DC}, U_{AC}), **gas composition** (air/N_2/O_2), **humidity** (RH%), **CPSI** on discharge properties, its stability, propagation velocity and chemical activity were briefly presented.
- Discharge propagation velocity in tubes was of ~ 10^7-10^8 cm/s and it ↑, for tube diameter ↓, oxygen content ↑ and negative polarity.
- Onset and breakdown voltages, and the amplitude of current pulses ↑ with tube diameter ↓.
- The discharge in tubes can be maintained by auxiliary discharge → effectively enhanced/suppressed by AC bias.
- Discharges in confined geometries generate a plasma that can be potentially used in various application utilizing plasma-catalytic system.