THE CONVERSION OF CARBON DIOXIDE IN RADIO-FREQUENCY DRIVEN ATMOSPHERIC PLASMA JETS

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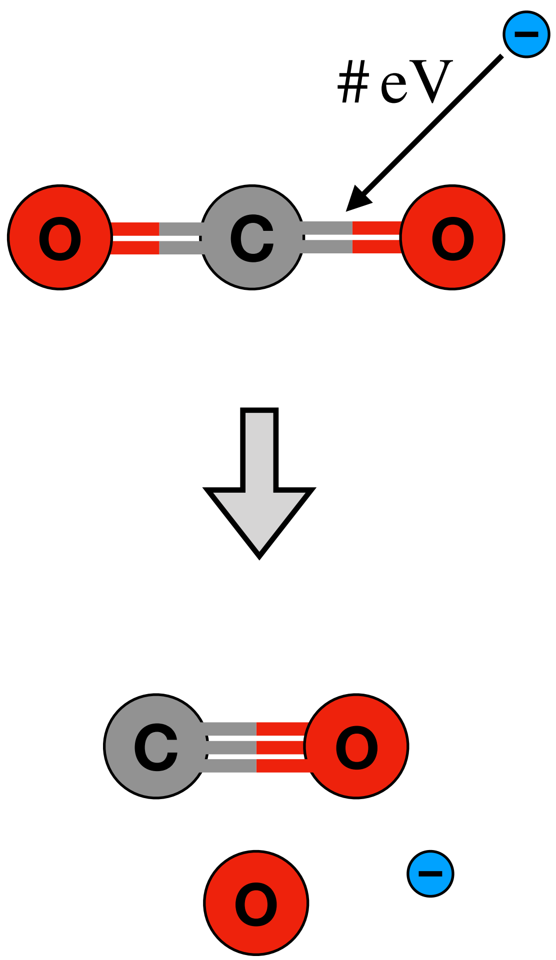
Carbon Dioxide Conversion

- CO_2 strongly contributes to an increase of greenhouse gases
- recycling into valuable chemicals and new fuels
- energy efficient splitting of CO_2
- plasma based conversion can replace thermal conversion
- using renewable energy sources
- energetic electrons lead to gas activation such as dissociation, ionization and excitation
- what kind of plasmas are suitable:
  - dielectric barrier discharge
  - microwave plasma
  - gliding arc discharge
  - radio-frequency driven plasma jets

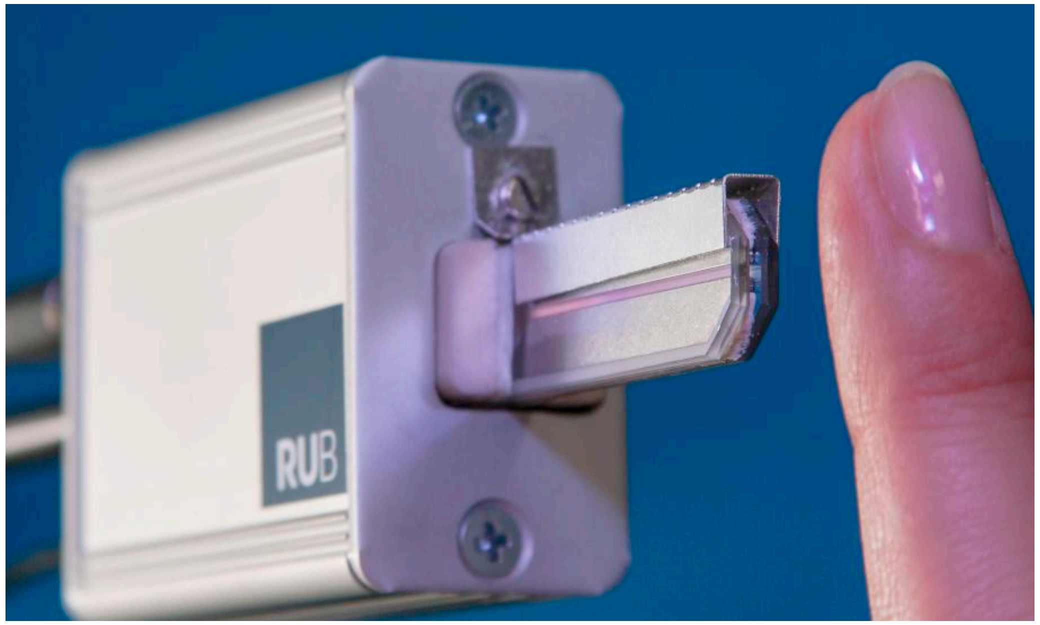
← **focus of this talk**



[1] Snoeckx and Bogaerts, Chemical Society Reviews 46. 19 (2017)

[2] Bogaerts and Centi, Frontiers in Energy Research 8 (2020)

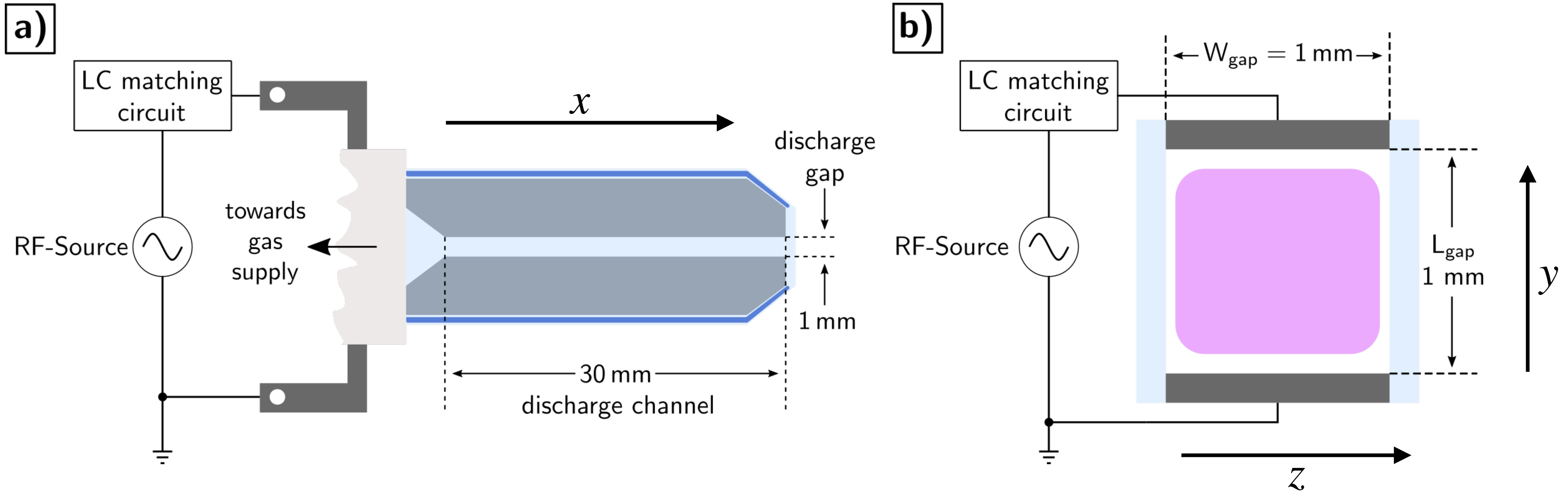
COST Reference Microplasma Jet



[3] Source: https://www.cost-jet.eu/

- based on the design of the μ-APPJ
- applications: water treatment, surface modification, biological applications, conversion of molecules
- radio-frequency driven (13.56 MHz, VWT)
- gas flow and mixture into a small discharge channel
- quadratic cross section of the channel (1x1 mm)
- 30 mm long channel reaching the effluent

→ **control of reactive species**



[3] Klich, Wilczek, Donkó and Brinkmann, Plasma Sources Sci. Technol. 31 045003 (2022)

How to Investigate this Process?

Chemistry Set

Reduced chemistry set

for He/CO_2

57 Species 354 Reactions

[4a] Lowke, Phelps, *J. Appl. Phys.* 44 4664–71 (1973)

[4b] Kozak, Bogaerts, PSST 23, 045004 (2014)

Global Models

0d or 1d plug flow model (globalKin):

Solving → Species balance equation
→ Electron energy equation

Fast simulation, suitable for investigating large parameter ranges

[5] Dorai, Kushner J. Phys. D: Appl. Phys. 35 2954 (2002)

Goal

- CO_2 conversion
- fundamental research
- electron dynamics
- validation

Fluid and Kinetic Models

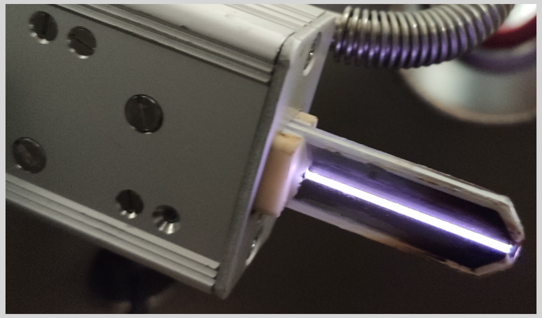
2d plasma fluid model (nonPDPSIM):

Hydrodynamics + Boltzmann solver

1d kinetic/hybrid models (PIC/MCC):

Particle based kinetic simulation in order to capture kinetic effects

Experimental Results



PROES Mass spectroscopy TDLAS

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2d Simulation: nonPDPSIM

Charged Species

$$\frac{dn_s}{dt} + \nabla \cdot \vec{\Gamma}_s = S$$

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

$$\frac{3}{2}\frac{d(n_s k_b T_s)}{dt} + \vec{E} \cdot \vec{j}_s + \nabla \cdot \vec{q}_s = \varepsilon_s$$

+

$$\nabla^2 \Phi = -\rho / \varepsilon_0$$

↔

Neutral Transport

Navier-Stokes equations

↔

Boltzmann-Solver

Conditions:

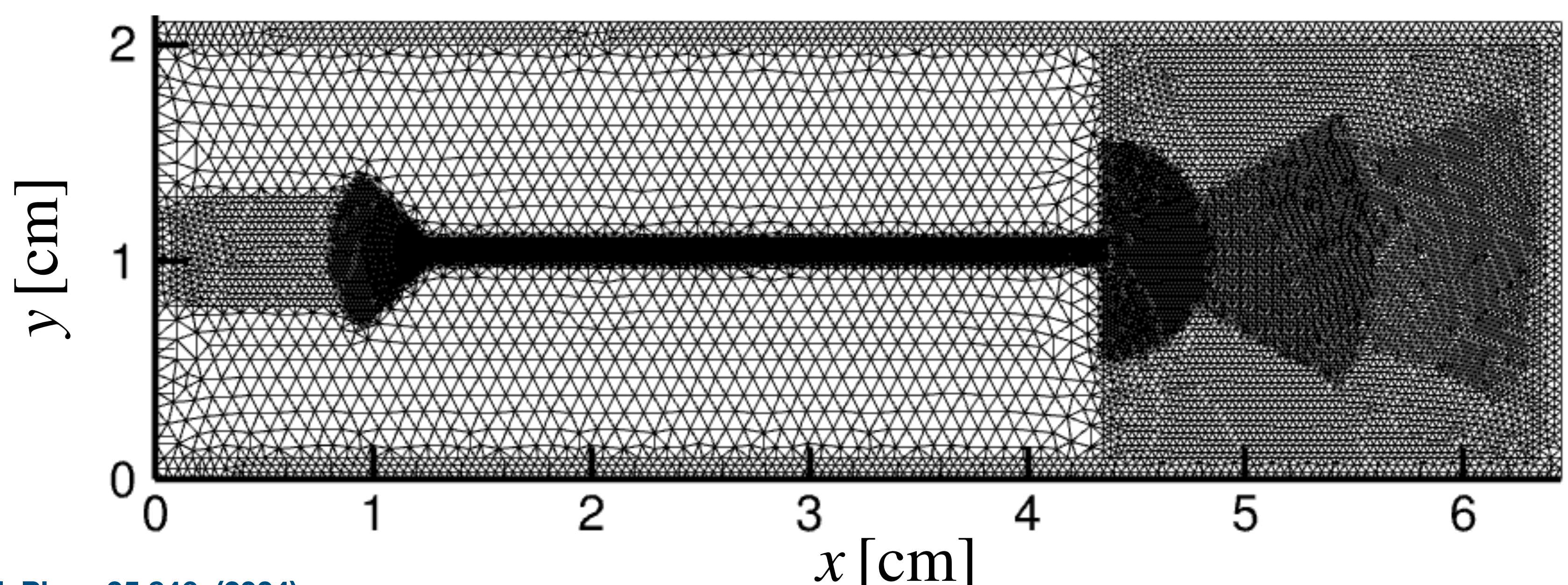
$550 \leq V_{\mathrm{RF}} \leq 1000\,\mathrm{V}$

$f = 13.56\,\mathrm{MHz}$

$p = 1\,\mathrm{atm}$

$\mathrm{He/CO_2} = 99.9/0.1$

flow rate $= 1000\,\mathrm{sccm}$

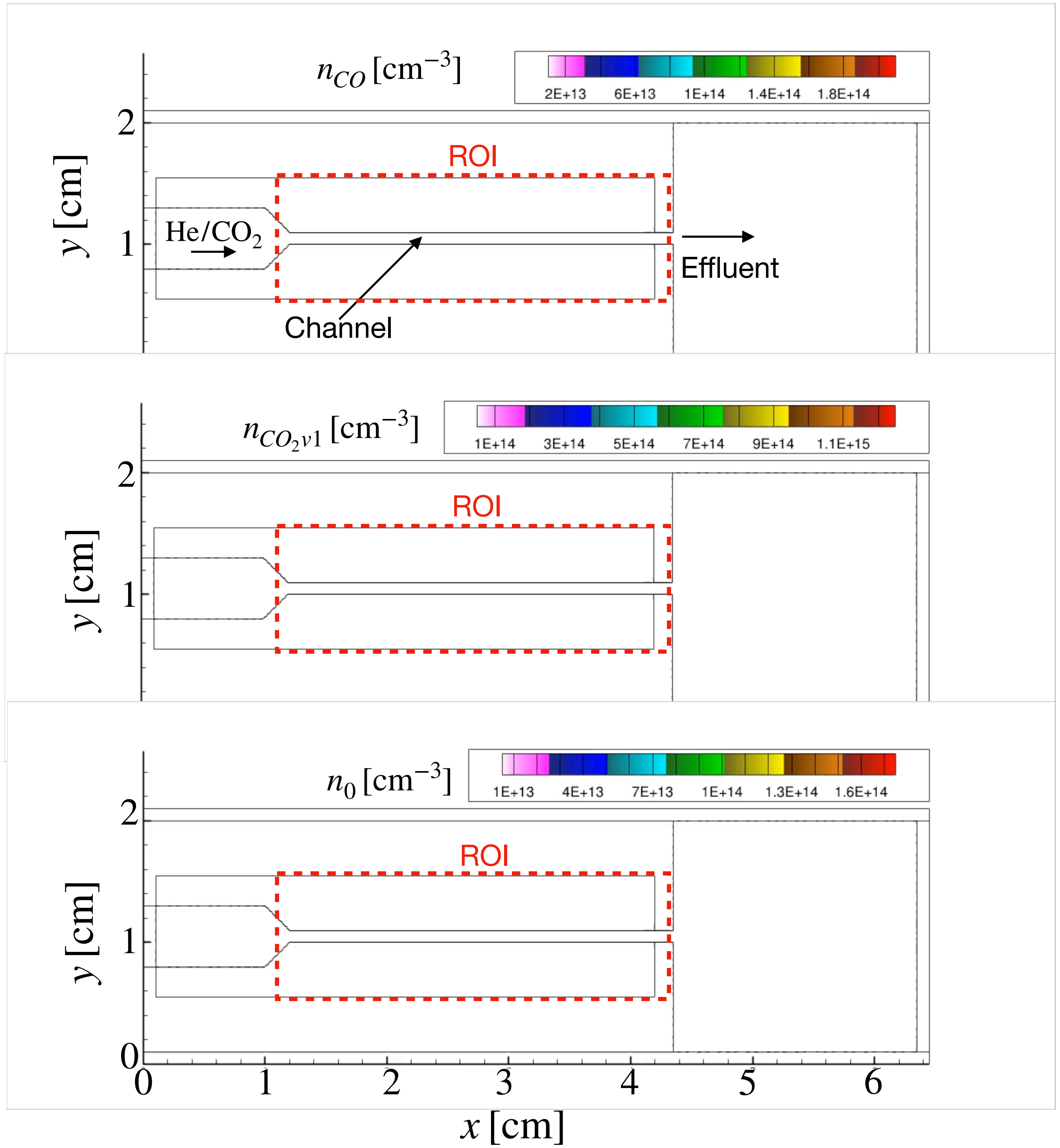


unstructured mesh for the 2d setup

[6] M J Kushner J. Appl. Phys. 95 846. (2004)

[7] M J Kushner J. Phys. D: Appl. Phys. 38 163 (2005)

Gas Dynamics



Conditions: $f = 13.56\,\mathrm{MHz}$ $V_{RF} = 650\,\mathrm{V}$ $p = 1\,\mathrm{atm}$
$\mathrm{He/CO_2} = 99.9/0.1$ $\text{flow rate} = 1000\,\mathrm{sccm}$

time scale of the effluent (ms)

- application: interaction of the effluent with materials and surfaces (water treatment)
- focus on CO_2 conversion
- region of interest (ROI) for the conversion is the discharge channel

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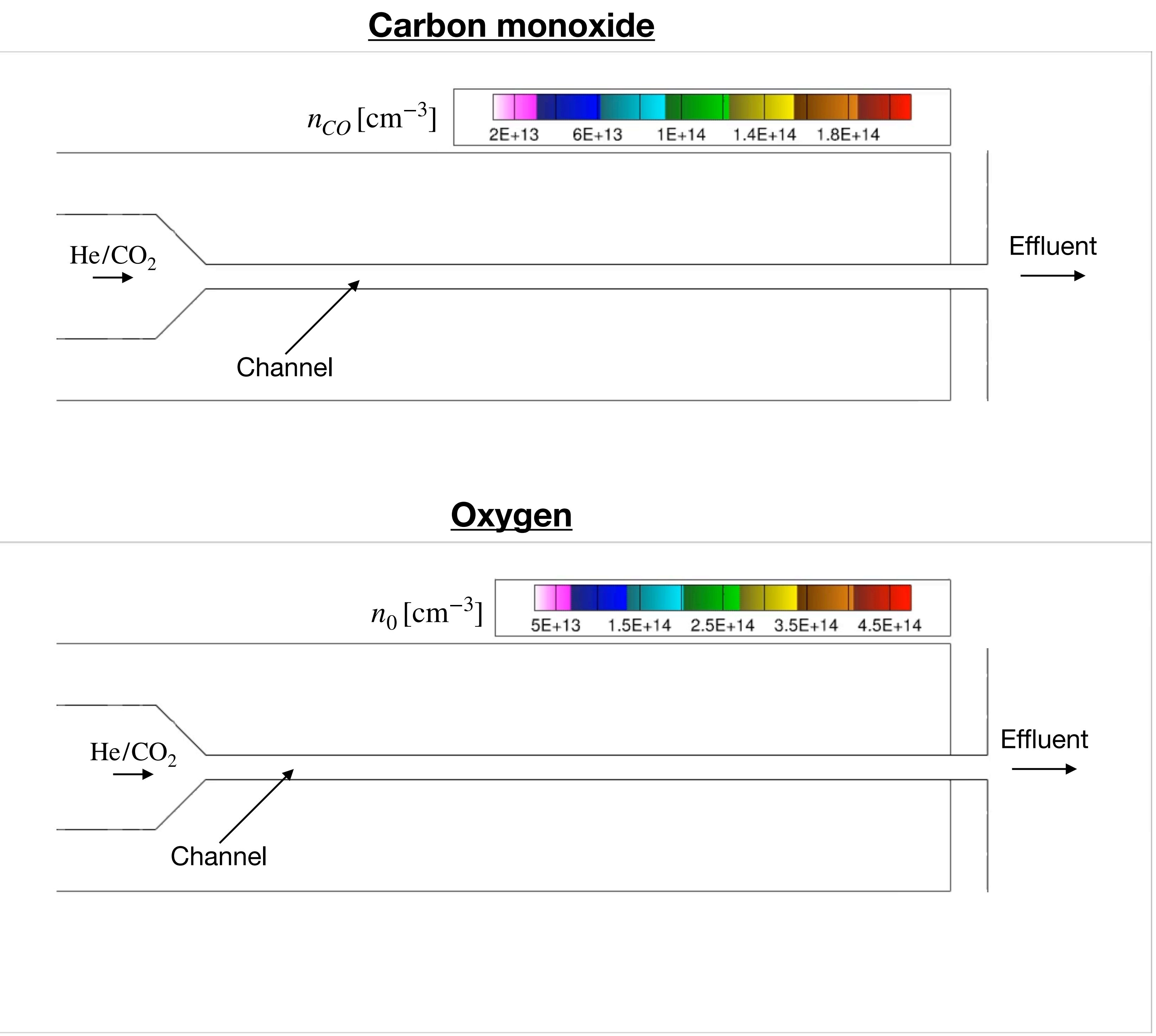
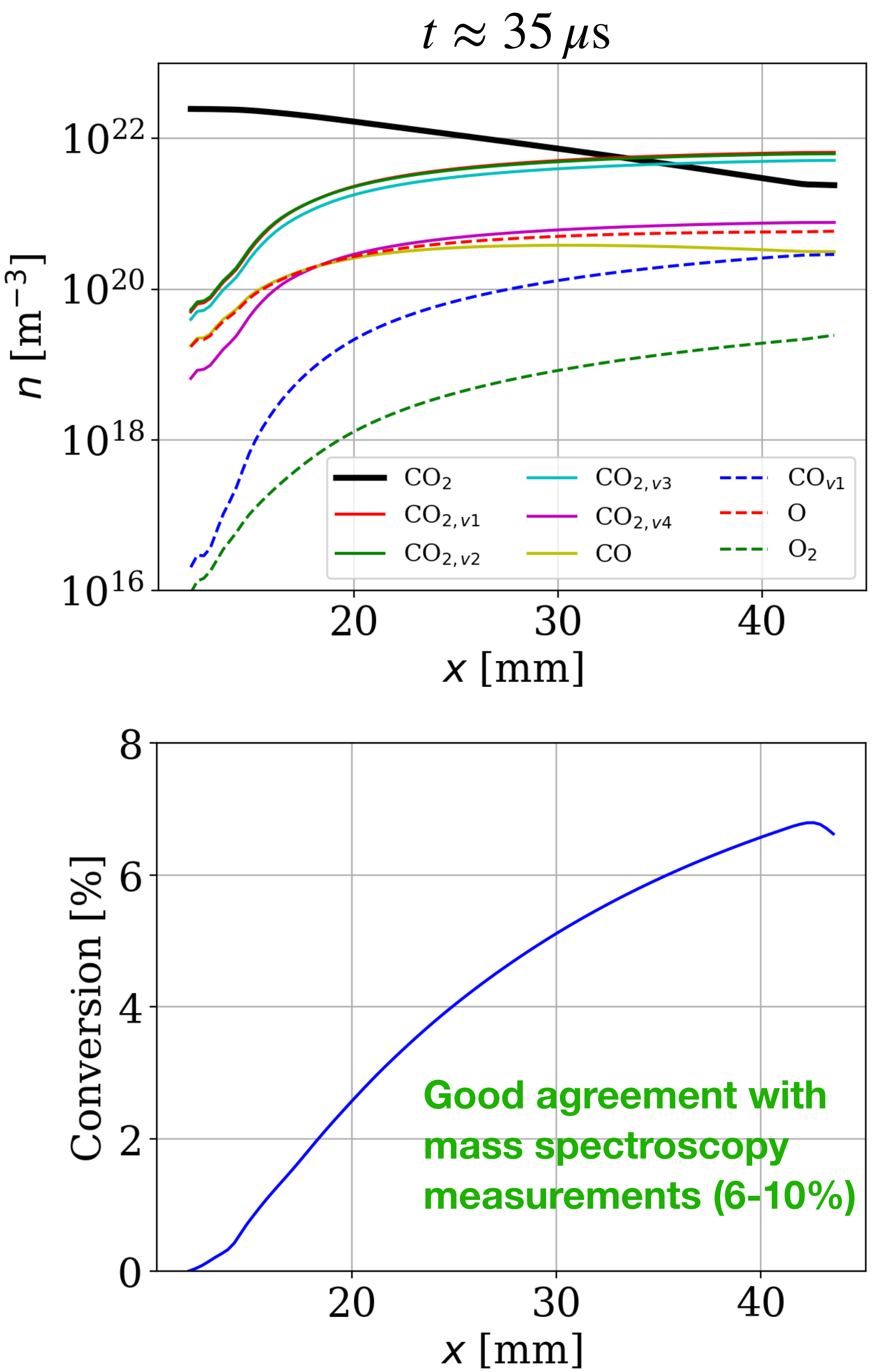
Conversion of Carbon Dioxide

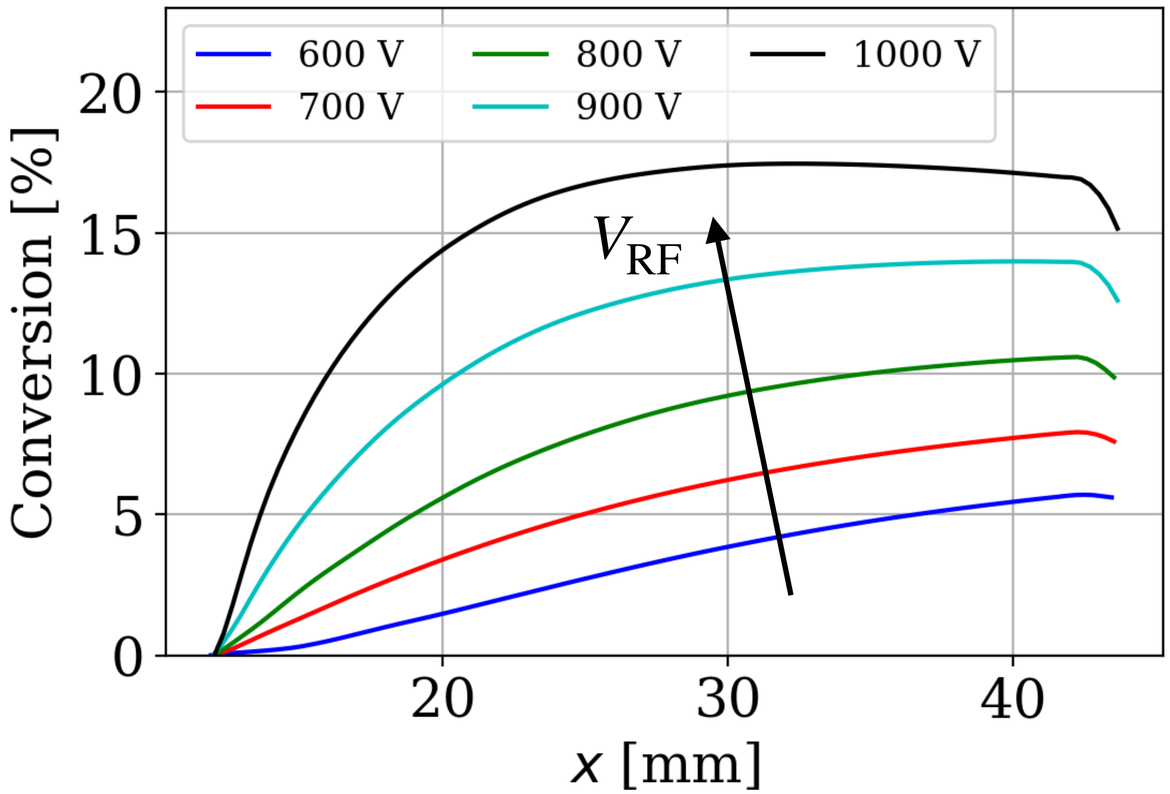




Conditions: $f = 13.56$ MHz $V_{RF} = 650$ V $p = 1$ atm
$He/CO_2 = 99.9/0.1$ flow rate $= 1000$ sccm

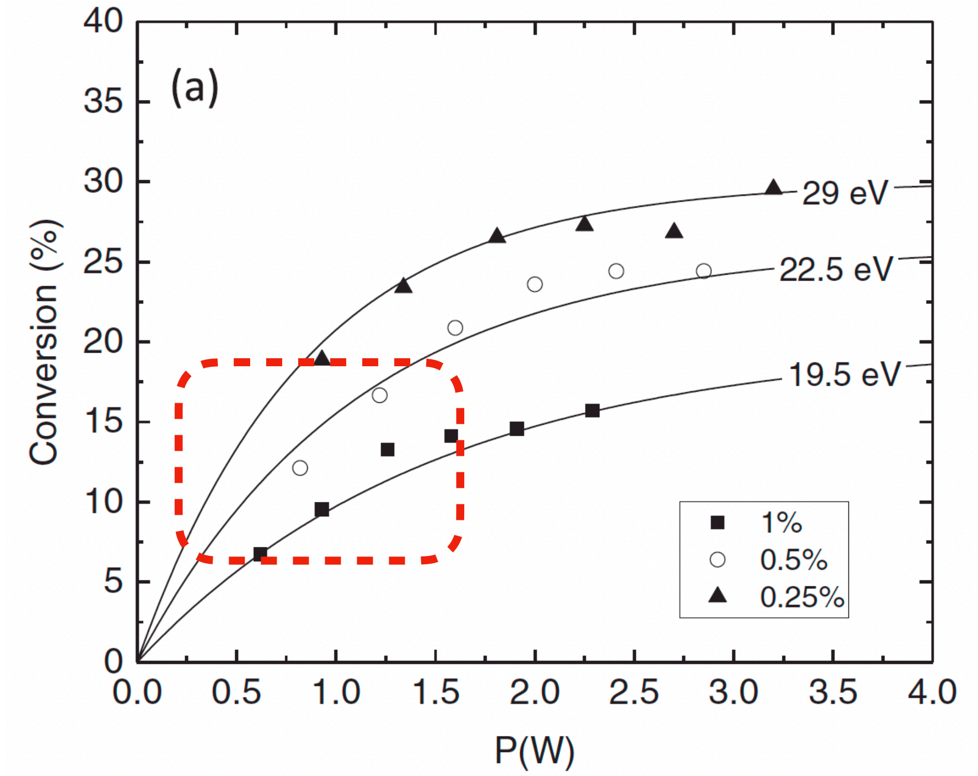
Conversion of Carbon Dioxide

COST-Jet (1 mm width)



Different-Jet (14 mm width)

[8] Urbanietz et al. J. Phys. D: Appl. Phys. 51 345202 (2018)

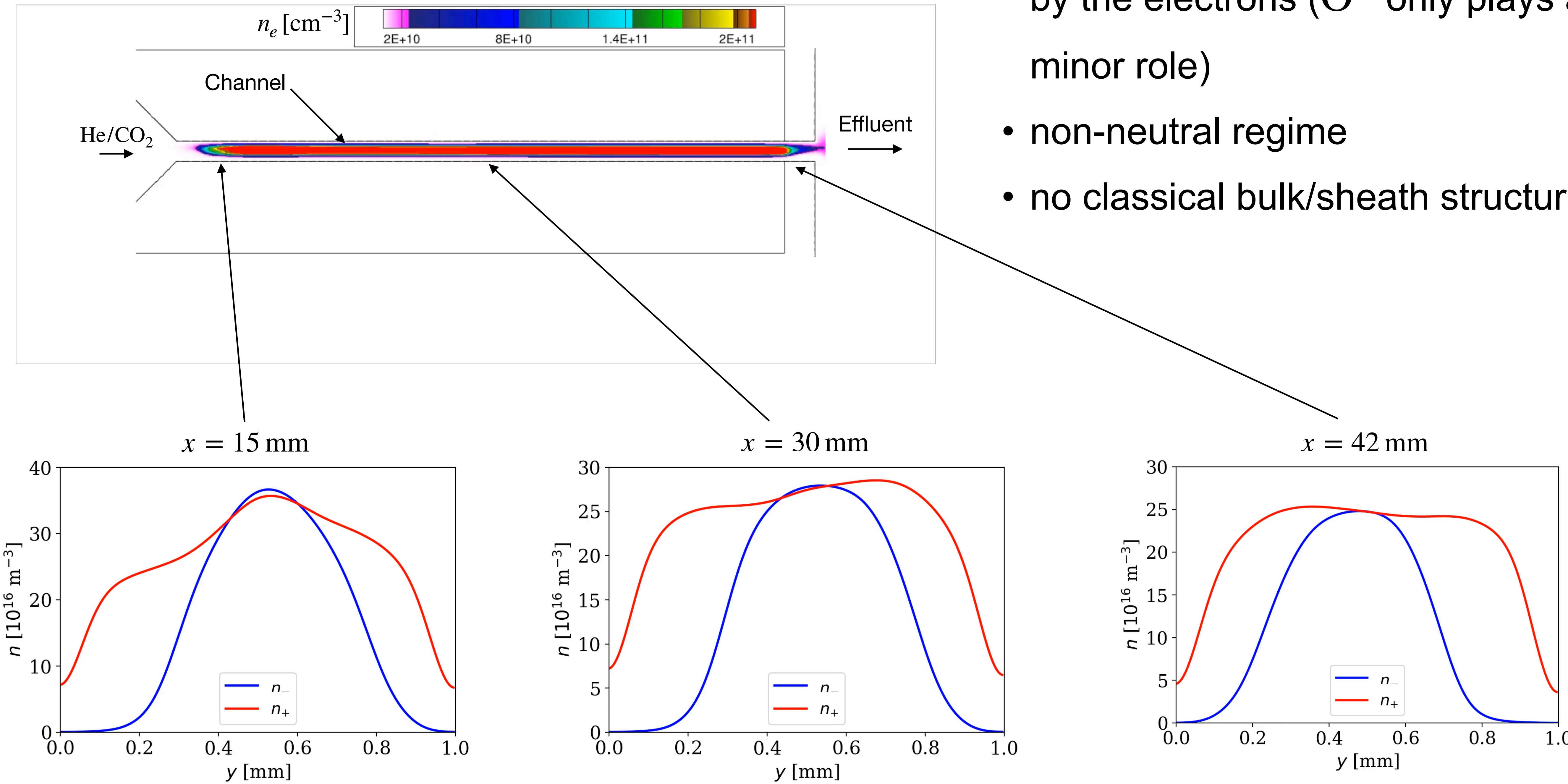


- increasing the RF voltage leads to higher conversion rate ($P \approx 1\,\mathrm{W}$)
- comparing this with similar experimental results (different jet design with 13 mm width using FTIR), a higher conversion can be achieved
- however, voltage from the simulation ($550 \leq V_{\mathrm{RF}} \leq 1000\,\mathrm{V}$) does not match correctly with experimental results ($200 \leq V_{\mathrm{RF}} \leq 700\,\mathrm{V}$)

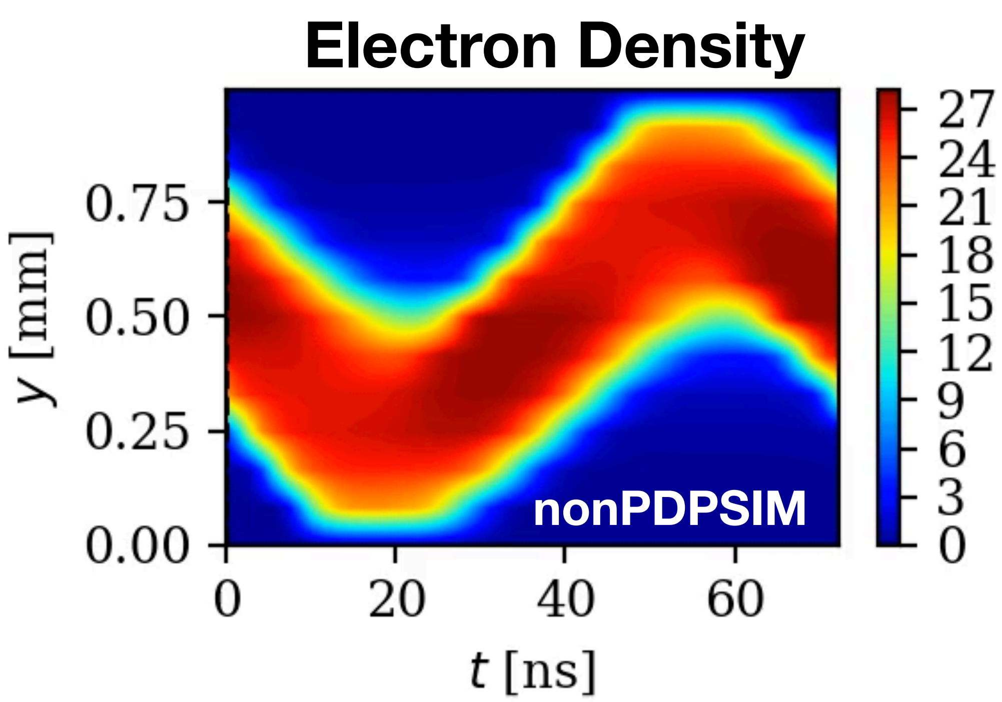
Electron Dynamics

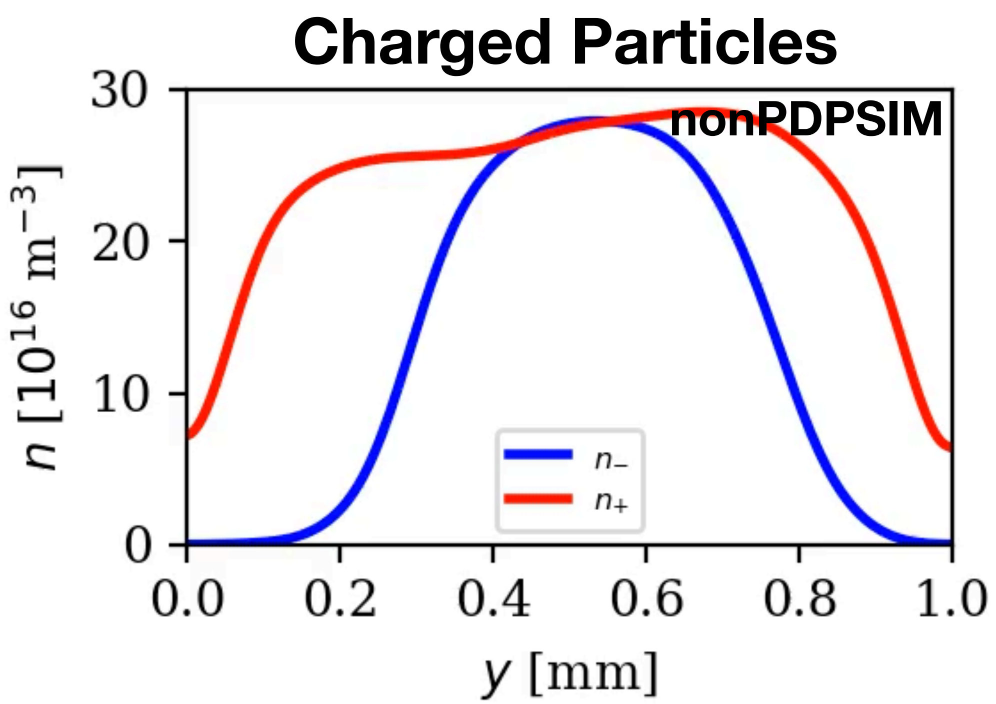
times scale of one RF-cycle: $T \approx 74\,\mathrm{ns}$

- dominant ions are O^+
- negative species are dominated by the electrons (O^- only plays a minor role)
- non-neutral regime
- no classical bulk/sheath structure



Comparison with PIC/MCC





nonPDPSIM

$f = 13.56\,\mathrm{MHz}$

$V_{RF} = 650\,\mathrm{V}$

$p = 1\,\mathrm{atm}$

$He/CO_2 = 99.9/0.1$

[3] Klich, Wilczek, Donkó and Brinkmann, Plasma Sources Sci. Technol. 31 045003 (2022)

[9] Vass, Wilczek, Schulze, Donkó, Plasma Sources Sci. Technol. 30 105010 (2022)

[10] S. Wilczek et al., Phys. Plasma 23, 063514 (2016)

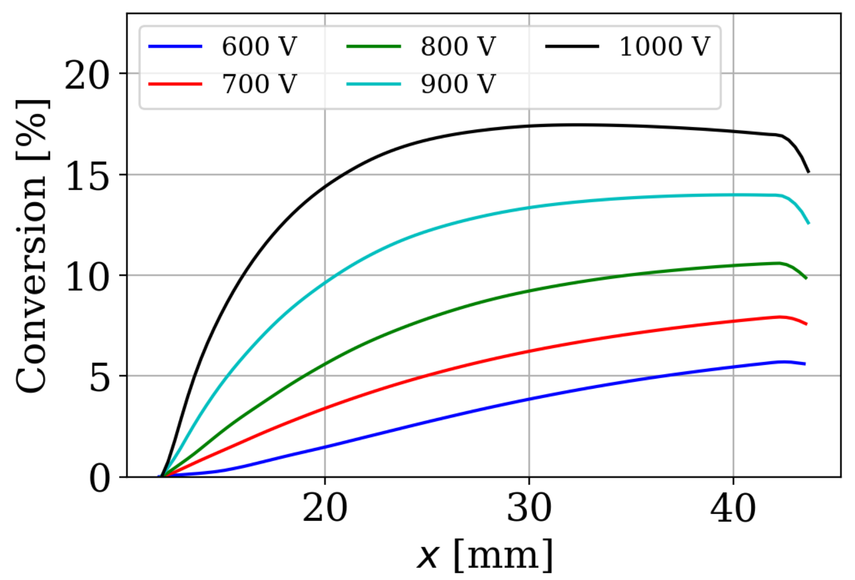
Summary and Outlook

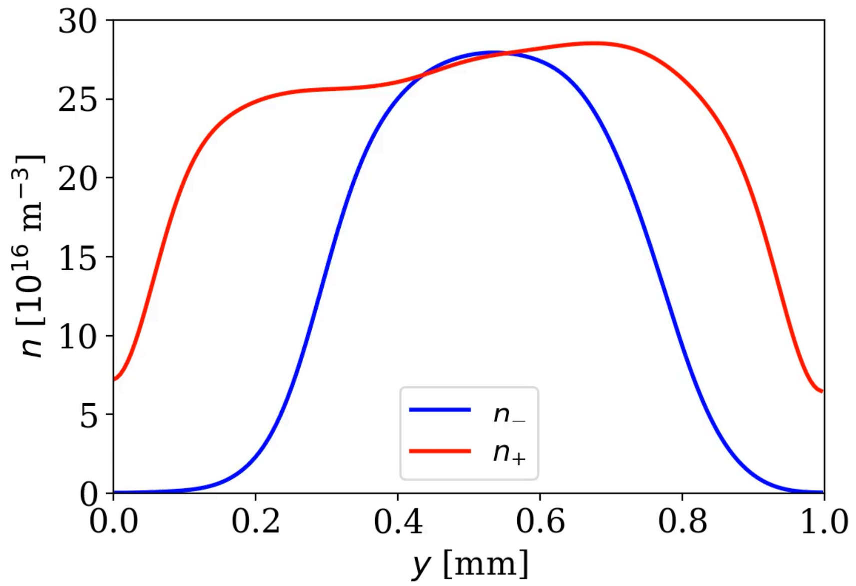
Summary

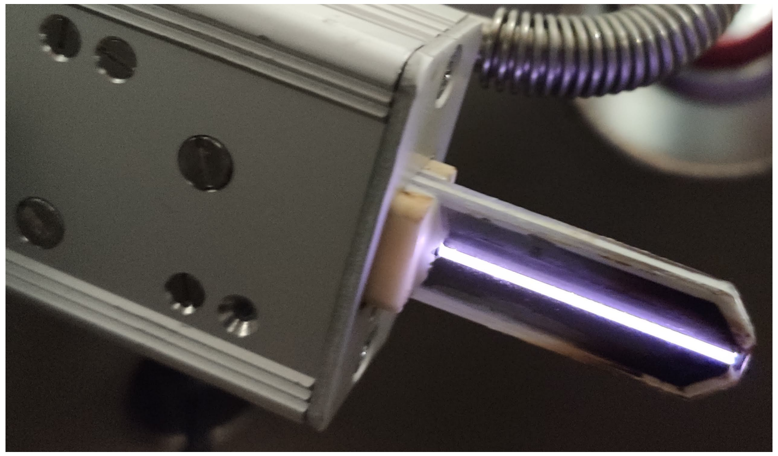
- CO_2 conversion was studied in the COST jet by 2d fluid simulations (nonPDPSIM)
- 18% conversion can be achieved by changing the RF voltage in the simulation
- electron dynamics show non-neutral dynamics, which is also observed in kinetic PIC/MCC simulations

Outlook

- chemistry set must be modified in order to include a more accurate dissociation channel
- the parameter range will be adjusted (different flow rate, higher driving frequencies, voltage waveform tailoring)
- experimental results (PROES, mass spectroscopy, TDLAS) will provide better insight about the potential operating parameters







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