DC glow discharge - Fluidized bed reactor for CO_2 recycling

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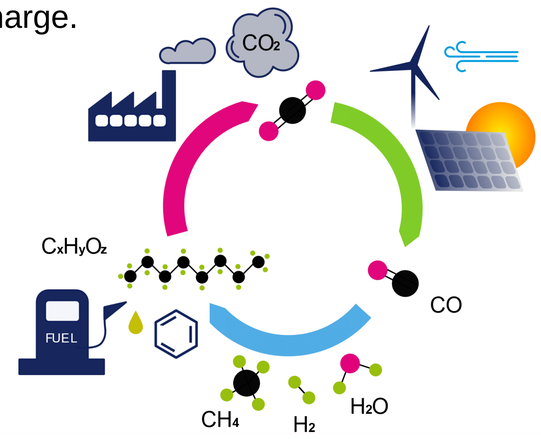
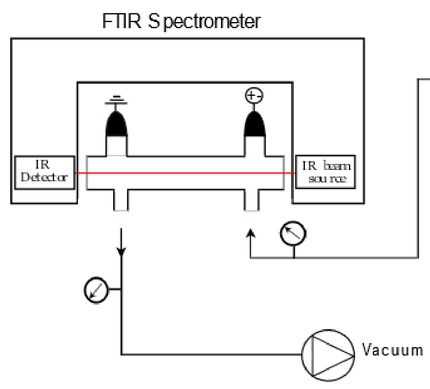
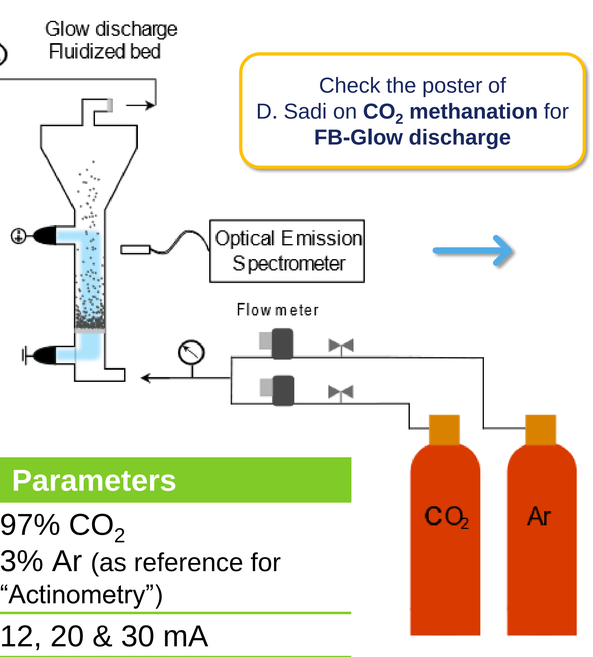
Using Plasma-Catalysis for CO_2 recycling

Closing the carbon cycle can be achieved by converting CO_2 into platform molecules or short hydrocarbons. Non-thermal plasmas can provide a peculiar environment out of equilibrium allowing CO_2 conversion with minimal energy cost, but they are poorly selective. The presence of a catalyst could greatly improve the conversion although the coupling is critical. **Fluidization** of the powder catalyst can increase the surface contact area with the plasma and improve the heat transfer. Fluidized bed reactors with plasma have proven an enhancement of conversion and a significant reduction in carbon deposition[1-3]. The development of this **innovative route** is crucial to understanding the enhancement of plasma-catalyst interaction for CO_2 recycling. In this work a fundamental study of plasma coupling with fluidized particle is performed in a low pressure glow discharge.

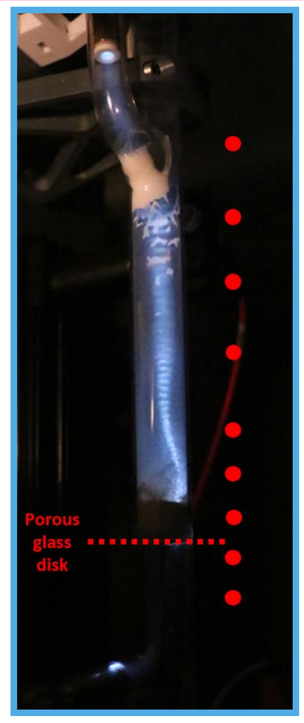


OES to characterize *in situ* glow discharge plasmas





Check the poster of D. Sadi on **CO_2 methanation** for **FB-Glow discharge**



CO Angström system

$B^3\Sigma^+ \rightarrow A^1\Pi$

CO 3rd Positive system (3PS)

$B^3\Sigma^+ \rightarrow a^3\Pi$

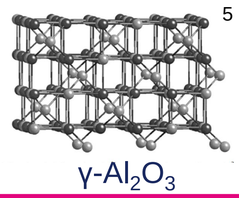
O atom emission lines

$777\ nm: {}^5P_{1,2,3} \rightarrow {}^3S$

$844\ nm: {}^3P_{0,1,2} \rightarrow {}^5S$

Parameters	
Gas	97% CO_2 3% Ar (as reference for "Actinometry")
Current	12, 20 & 30 mA
Pressure	1.5, 2 & 2.5 torr (positive column)
Flowrate	20 sccm
Material	Al_2O_3 (~100 μm size)
Quantity	0 (empty reactor) & 4 g

- ✓ High mechanical strength
- ✓ High surface area
- ✓ Inexpensive



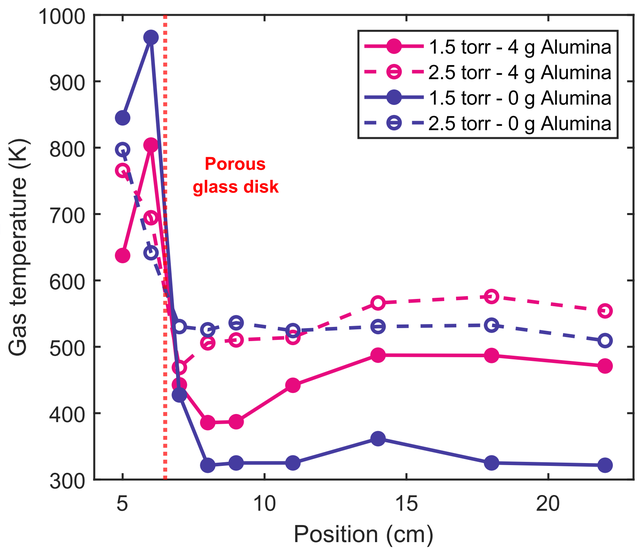
γ-Al_2O_3

The gas temperature does not increase significantly

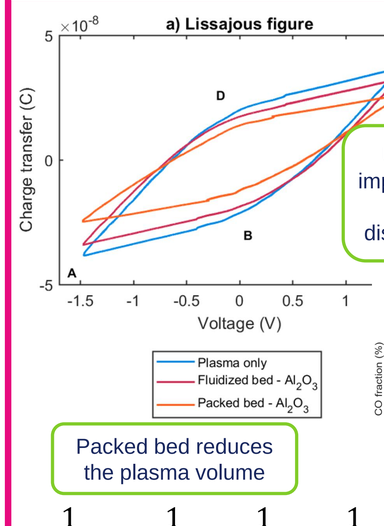
$T_{rot\ (CO)}$

$(v_1,v_2) = (0,1)$ optical transition of CO Angstrom band[4]

$T_{rot\ (CO)} \approx T_{gas}$

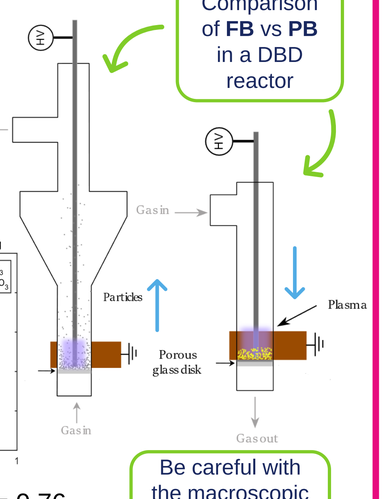


Fluidized bed vs Packed bed?



Unclear improvement on the dissociation

Comparison of **FB** vs **PB** in a DBD reactor



Packed bed reduces the plasma volume

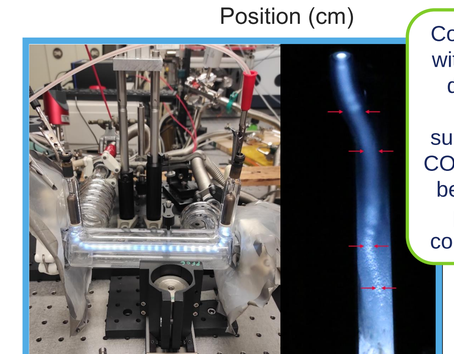
$$\frac{1}{C_{cell}} = \frac{1}{die1} + \frac{1}{gas} + \frac{1}{die2}$$

PB = 0,76
FB = 0,95

Be careful with the macroscopic properties of catalysts!

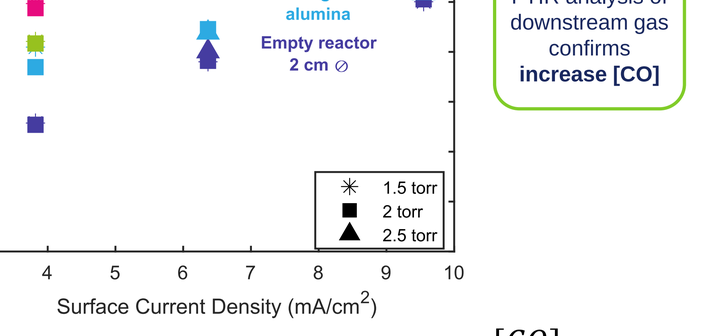
Movement of Al_2O_3 particles affects less the ignition of filaments and therefore, the charge transfer

Can the fluidization of particles in the plasma help on CO_2 dissociation?



Increase surface area: Particles help to recombine O into O_2

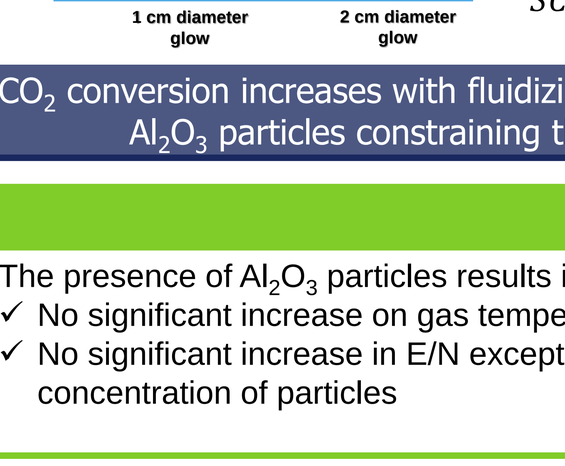
$$\frac{I_O^{777\ nm}}{I_O^{844\ nm}} \approx \frac{k^{777\ nm}}{k^{844\ nm}}\left(\frac{E}{N}\right) \rightarrow \downarrow f\left(\frac{E}{N}\right)$$



LoKI simulation on EEDF for CO_2 plasma

Increase of **E/N** in high particle density region

What about CO?



Similar trend with CO 3PS

Reduction of **E/N** towards the exit of the reactor with Al_2O_3 particles

$$I_{co} \sim [CO], n_e, k\left(\frac{E}{N}\right)$$

Increase in CO intensity is related to higher CO density

Comparison with smaller diameter reactor suggest that CO increases because of plasma confinement

1 cm diameter glow | 2 cm diameter glow

FTIR analysis of downstream gas confirms **increase [CO]**

$$SCD = \frac{Current\ (mA)}{Area\ (cm^2)} \qquad Conv.CO_2\ (\%) = \frac{[CO]_{out}}{[CO_2]_{in}} * 100$$

CO_2 conversion increases with fluidizing particles in the plasma region probably as a result of Al_2O_3 particles constraining the plasma spatially (i.e. higher current density)

Conclusions

The presence of Al_2O_3 particles results in:

- ✓ No significant increase on gas temperature
- ✓ No significant increase in E/N except at high concentration of particles
- ✓ Notorious increment in CO_2 conversion due to the spatial constrain of the plasma
- ✓ No clear improvement in dissociation but fluidized bed affects less the ignition of filaments in comparison to packed bed

References:
[1] Wang et al., Catal. Today, **148**, pp. 275–282 (2009)
[2] Chen et al. J. CO2 Utilization, **54**, p. 101771 (2021)
[3] Pou et al, J. CO2 Utilization, **27**, pp. 528–535 (2018)
[4] Yamada et al, Jpn. J. Appl. Phys., **60** (2021)
[5] Prins, J. of Catalysis, **392** pp. 336-346 (2020)

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PIONEER PLASMA CATALYSIS CO_2 RECYCLING

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