

# Non thermal plasma-assisted catalytic oxidation of methane: effect of catalysts.

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## Introduction

- ❑ Agriculture accounts to 10% of total green house gases in EU [1].
- ❑ Methane contributes 54% of these emission which is produced by rumination and belching by cattle [1].
- ❑ Methane, a potent greenhouse gas, has 28 times the global warming potential of CO<sub>2</sub> [2].
- ❑ Methane emissions in dairy barns and cattle farms are dispersed and occur in low concentration (20 – 200 ppm) [1, 2].
- ❑ Currently, no feasible technology to abate these low concentration of methane from dairy barns.
- ❑ **Goal of CANMILK project:** to capture methane emission from dairy barn and convert them into CO<sub>2</sub> using plasma and catalysis.
- ❑ Plasma, an ionized gas, triggers chemical reactions, but gas-phase plasma reactions are not selective [3].
- ❑ The use of oxidation catalysts can improve complete oxidation of methane into CO<sub>2</sub> [3].

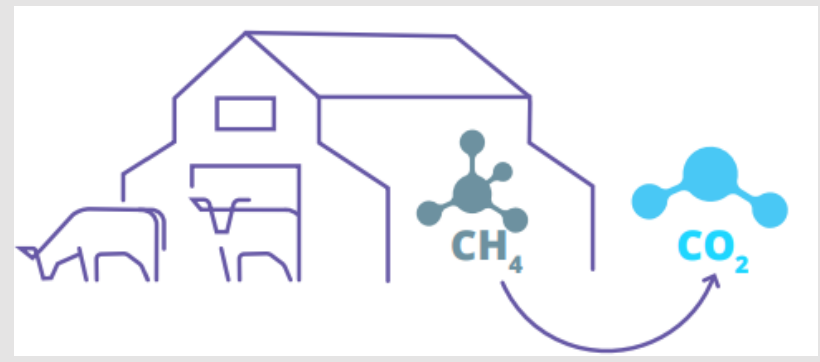


Figure 1. Goal of CANMILK project<sup>[1]</sup>.

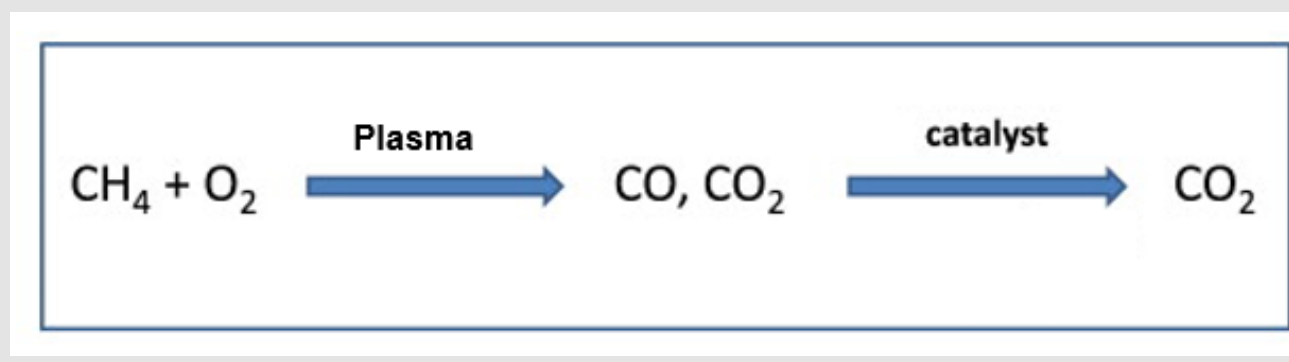


Figure 2. Plasma oxidation scheme<sup>[4]</sup>.

## Experimental

- ❑ 10 different catalysts composition: 1% Pd/Al<sub>2</sub>O<sub>3</sub>, 1% Cu/ Al<sub>2</sub>O<sub>3</sub>, 1% Pt/ Al<sub>2</sub>O<sub>3</sub>, 1% Co/ Al<sub>2</sub>O<sub>3</sub>\*, 1%Fe/ Al<sub>2</sub>O<sub>3</sub>\*, 3% Pd/ Al<sub>2</sub>O<sub>3</sub>, 1% Pd – 0.5% Cu/ Al<sub>2</sub>O<sub>3</sub>, 1% Pd – 0.5% Co/ Al<sub>2</sub>O<sub>3</sub>\*, 1% Pd – 0.5% Fe/ Al<sub>2</sub>O<sub>3</sub>\*, and 1% Pd – 0.5% Pt/ Al<sub>2</sub>O<sub>3</sub>\*.
- ❑ Prepared by vacuum impregnation, calcined at 500 °C and 8 hour in air. Sequential impregnation for bimetallic catalyst.
- ❑ Catalysts are characterized by N<sub>2</sub> physisorption, SEM-EDS, and XRD.

\*Catalysts are prepared but not tested for activity yet.

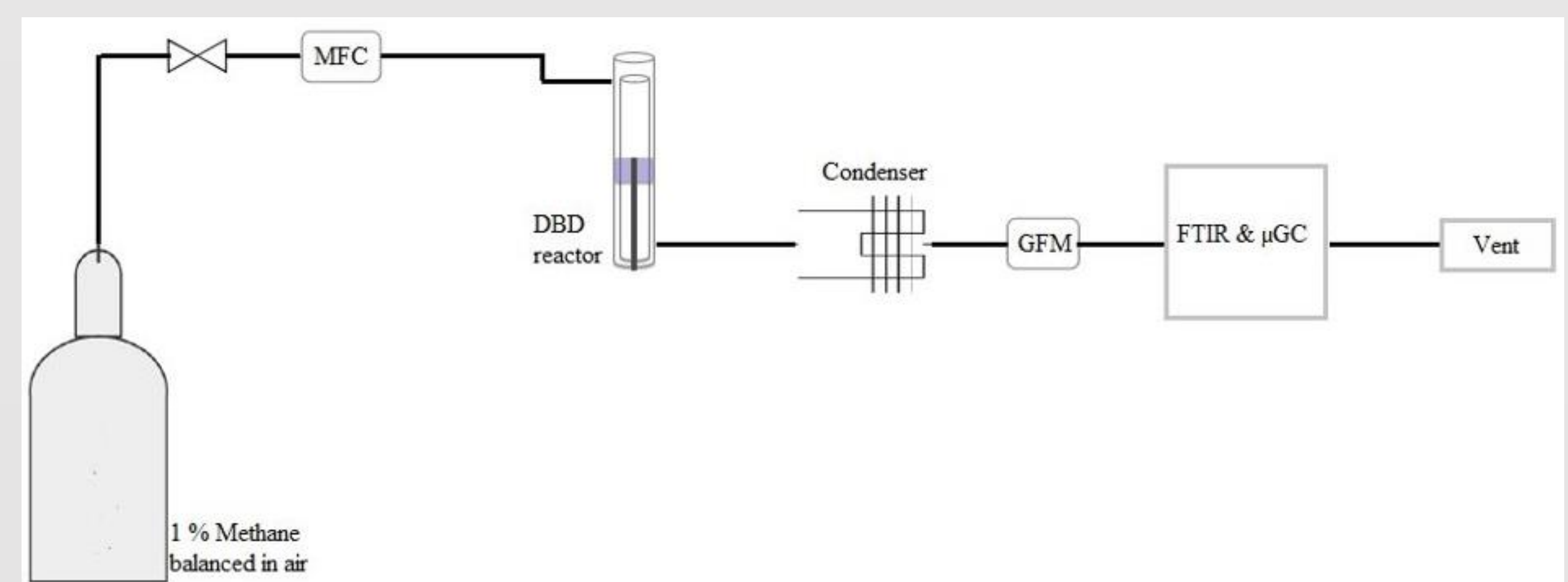


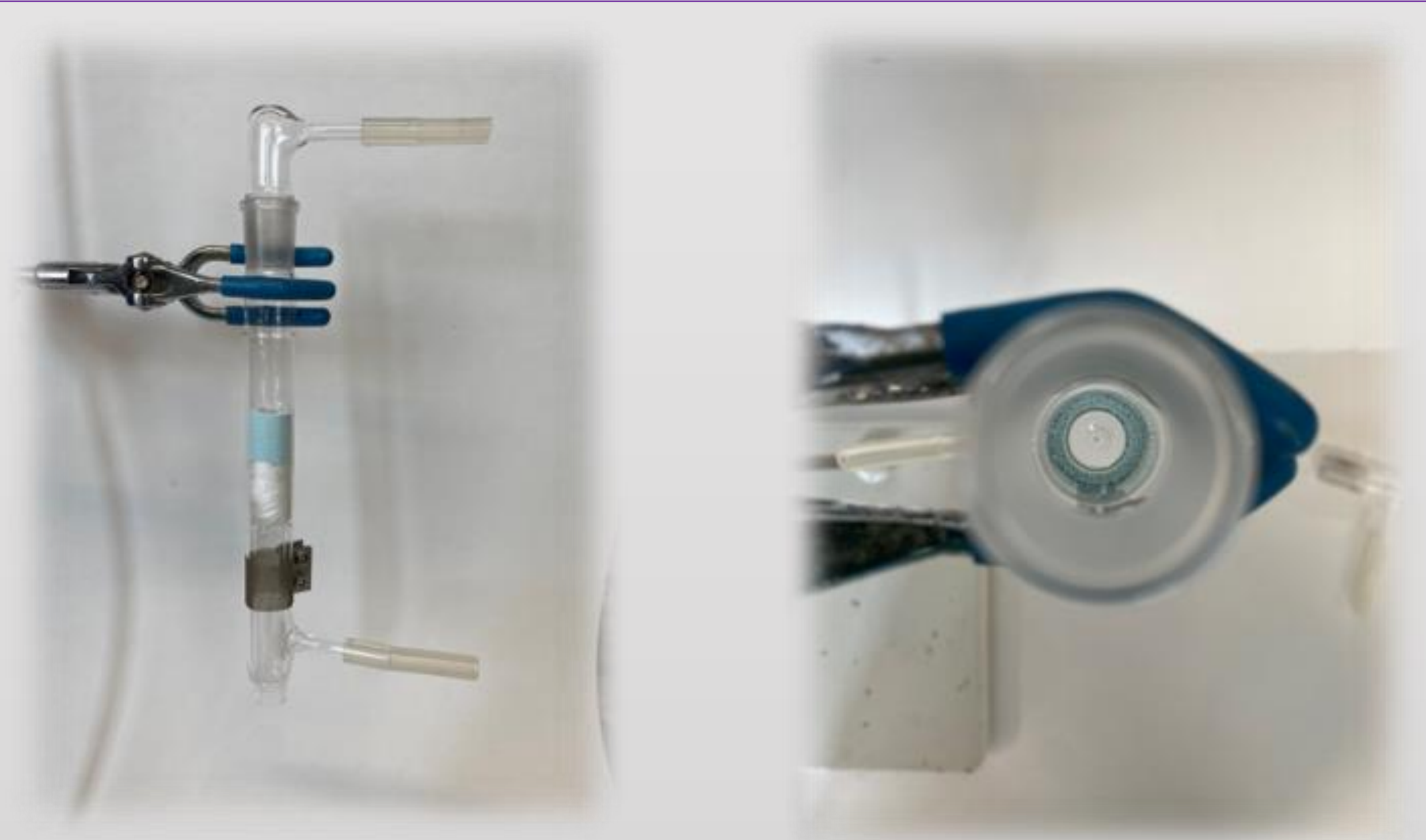
Figure 3. The experimental set up.

## DBD reactor

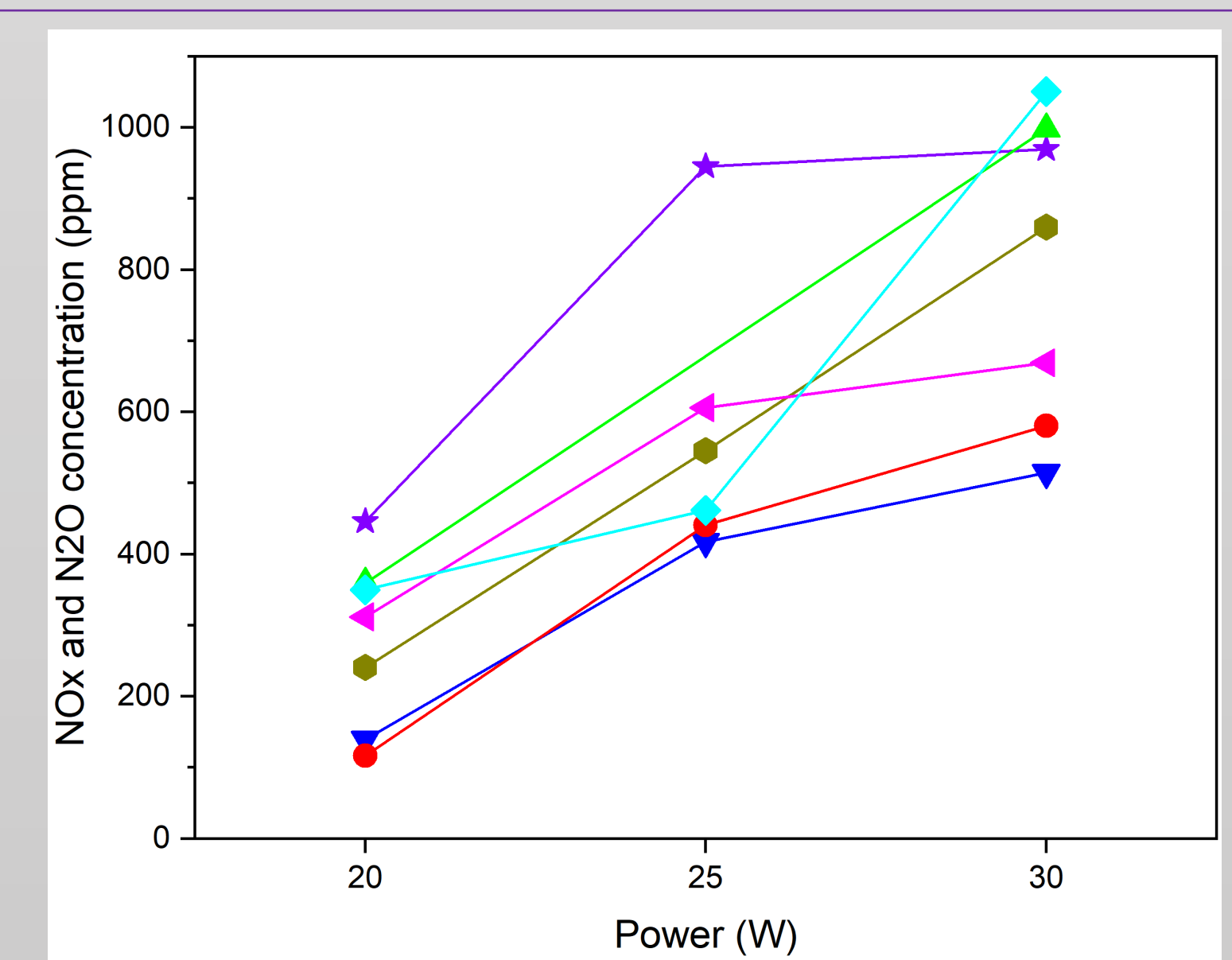
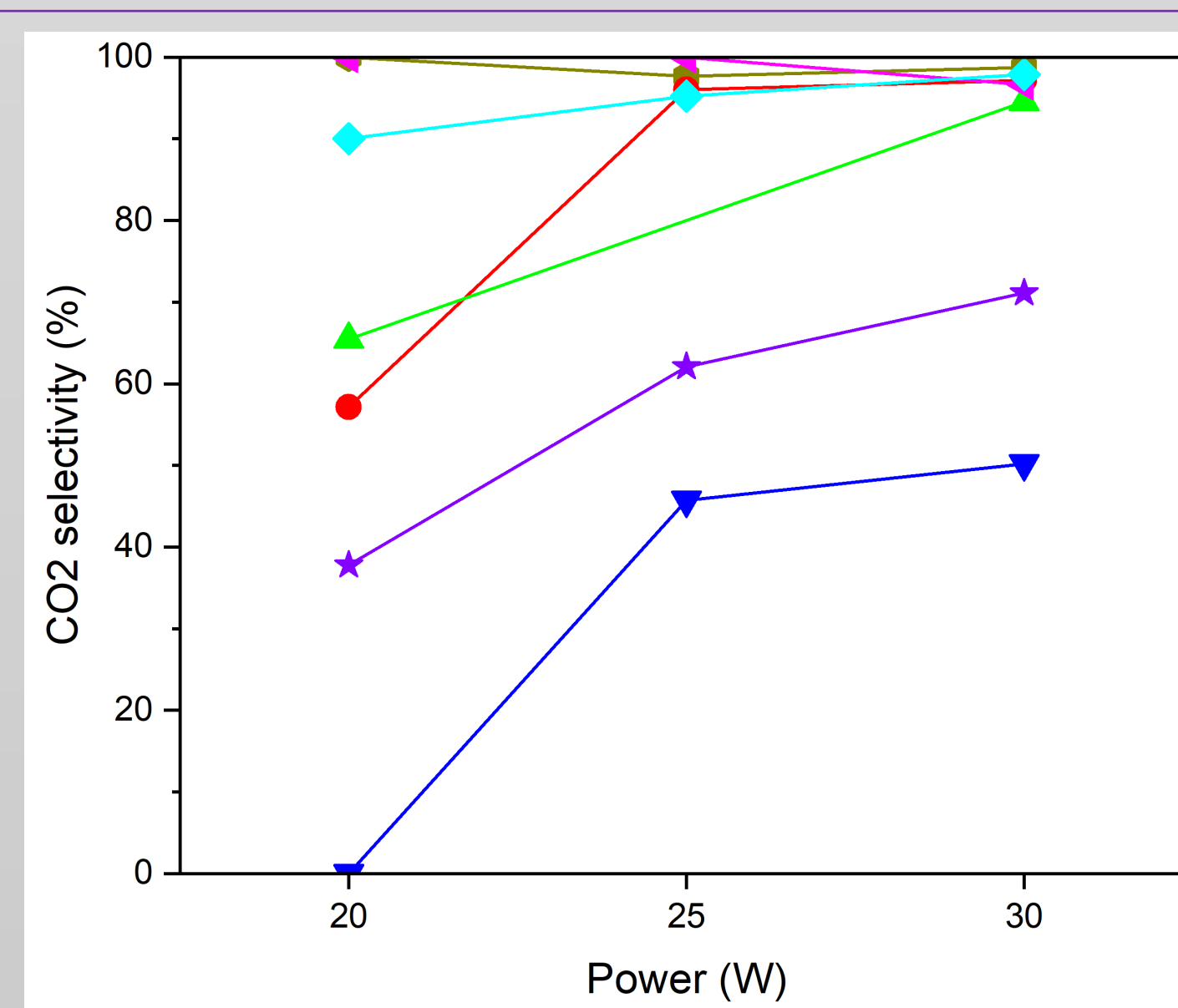
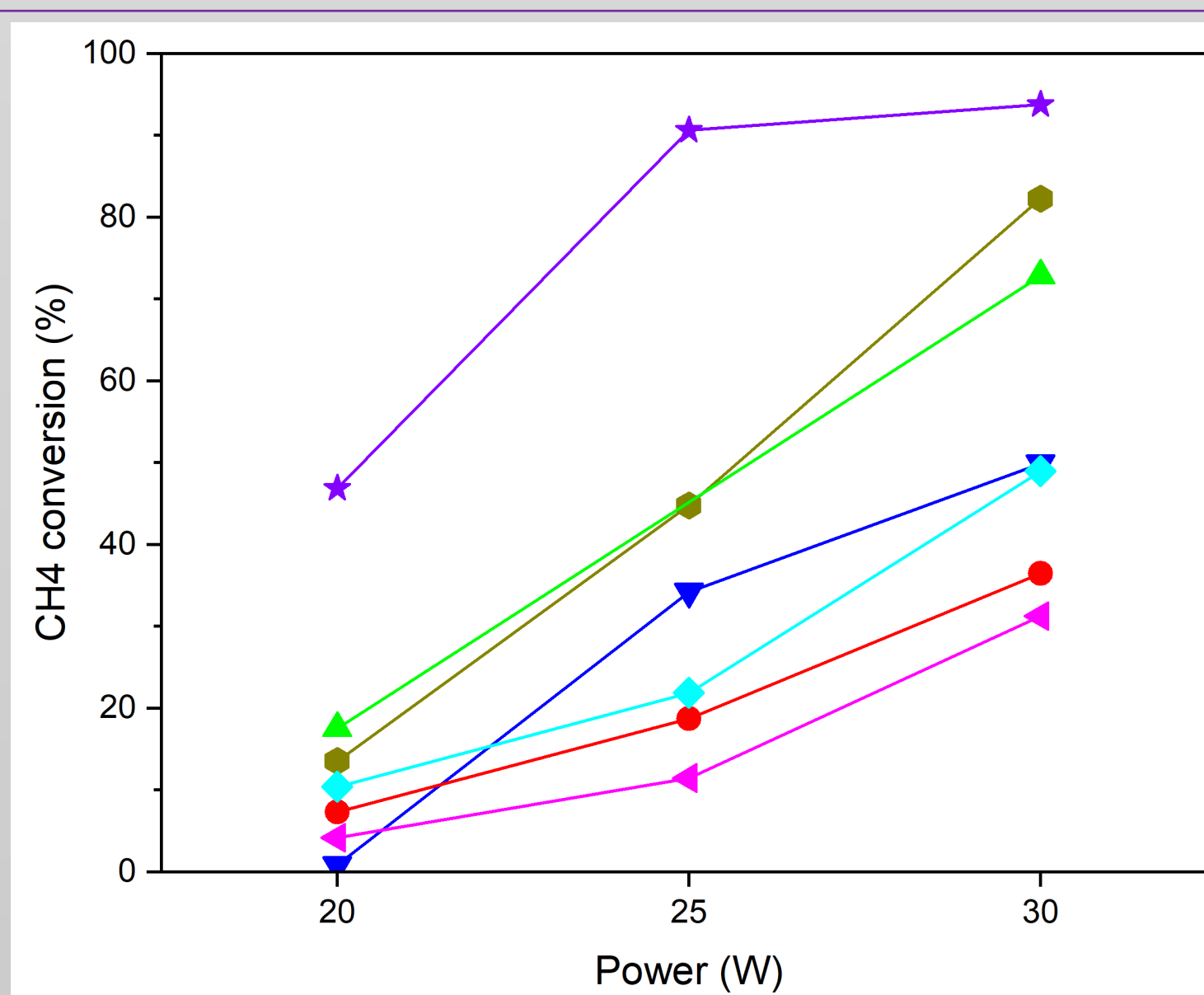
- ❑ Co-axial Dielectric barrier discharge (DBD) reactor.
- ❑ Two concentric quartz tubes.
- ❑ Discharge length: 2 cm and discharge gap: 2 mm.
- ❑ Stainless steel electrodes: Higher voltage inner electrode and outer ground electrode.
- ❑ Catalysts were placed inside the plasma discharge zone.



Figure 4. DBD reactor during experiment, catalyst loading inside a DBD reactor and top view of in-plasma catalyst system.



## Results



★ Plasma ▼ Al<sub>2</sub>O<sub>3</sub> ● 1% Pd/Al<sub>2</sub>O<sub>3</sub> ● 1% Pt/Al<sub>2</sub>O<sub>3</sub> ▲ 1% Cu/Al<sub>2</sub>O<sub>3</sub> ◆ 3% Pd/Al<sub>2</sub>O<sub>3</sub> ◆ 1% Pd-0.5% Cu/Al<sub>2</sub>O<sub>3</sub>

Figure 5. Effect of plasma power and catalyst on methane conversion, CO<sub>2</sub> selectivity and NO<sub>x</sub> and N<sub>2</sub>O formation. **Experimental conditions:** 200 mL/min (1% methane in air), atmospheric pressure and temperature

## Conclusions

- ❑ Plasma successfully activated methane, achieving a 90% conversion rate at 30 W plasma power.
- ❑ The CH<sub>4</sub> conversion increased with higher plasma power.
- ❑ The presence of catalyst reduced CH<sub>4</sub> conversion, but it resulted in complete oxidation of methane and improved CO<sub>2</sub> selectivity.
- ❑ 1% Pd/Al<sub>2</sub>O<sub>3</sub> catalyst showed 84% methane conversion and 99% CO<sub>2</sub> selectivity, showing promising signs for application in dairy barns and meat farms.

## References

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4. Lee H, Lim TH, Kim DH. Complementary effect of plasma-catalysis hybrid system on methane complete oxidation over non-PGM catalysts. Catal Commun. 2015 Jul 13;69:223–7.