CANMILK

GREENHOUSE GAS REDUCTION IN AGRICULTURE
USING PLASMA-BASED SOLUTIONS

DELIVERABLE D2.4

Oxygen production efficiency and CH_4 removal efficiency in microwave plasma for conditions selected on basis of D2.1

PROJECT GRANT AGREEMENT NO	DUE DATE OF DELIVERABLE:	ACTUAL SUBMISSION DATE:	START DATE OF PROJECT:	PROJECT DURATION:
101069491	31.01.2024	17.02.2024	01.09.2022	48 MONTHS

Funded by
the European Union

Work Package	WP2
Associated Task	D2.4
Deliverable Lead Partner	Gerard van Rooij
Main author(s)	Stijn Helsloot
Internal Reviewer(s)	Thomas Butterworth
Version	

DISSEMINATION LEVEL		
PU	Public	
PP	Restricted to other programme participants (including the Commission Services)	
RE	Restricted to a group specified by the consortium (including the Commission Services)	
CO	Confidential, only for members of the consortium (including the Commission Services)	

CHANGE CONTROL

DOCUMENT HISTORY

VERSION	DATE	CHANGE HISTORY	AUTHOR(S)	ORGANISATION
1.0	26-1-2024	1st Version	S.Helsloot	UM
2.0	31-1.2024	2nd Version	-	UM

This deliverable is part of a project that has received funding from the European Union's Horizon Europe research and innovation programme under grant agreement no 101069491.

EXECUTIVE SUMMARY

UM has built up an experimental microwave plasma system for destruction of enteric methane emissions. In this system, trace methane can be injected into the afterglow of either an O_2, H_2, or air plasma. To asses the performance of this reactor, we have developed a method for quantification of the reaction products using Fourier transform infrared (FTIR) spectroscopy. Quantification is done by fitting procedures that use both experimental and theoretical spectra. The measurements indicate that both air and O_2 yield promising results, being capable of converting trace methane via either thermal or radical conversion.

DEVIATIONS

- In our experimental programme, we tested the plasma forming gas in the following order: H_2, N_2, Air, and O_2. In the original plan, O_2 was listed first in the agenda, but we deviated due to safety challenges associated with pure O_2. This change of agenda prevented timely availability of O2 plasma data to support modelling activities in Antwerp.
- We started our experiments with Methane in Argon (0.1% CH_4, 99.9% Ar) instead of synthetic barn-air (0.24% CH_4, 20% O_2, and 79.76% N_2).
- Up to now, no laser-diagnostics have been possible. Due to this, we made the logical conclusion that it was smarter to start with the direct performance measurements of both Hydrogen and Oxygen. We also put in the extra effort by looking at N_2 and air.

TABLE OF CONTENT

LIST OF FIGURES

LIST OF ABBREVATIONS

ACRONYM	DESCRIPTION
D	Deliverable
EC	European Commission
WP	Work package
WT	Work task

1. INTRODUCTION

The original plan was as followed:

> *Microwave plasma, the workhorse within UM for its flexibility in operation and diagnostic access, is invoked to create a reference set of experimental results that can be used to validate the chemical modelling at UA (T2.1-T2.3). The initial experiments focus on formation of O-atoms and their subsequent consumption by traces of CH_4 that are supplied in the effluent. Absolute species concentrations are quantified with Raman. The downstream formation of hydrocarbons, OH and water is detected with in situ FTIR with the purpose to quantify the overall CH_4 removal efficiency. A research scientist from VTT will participate in the work.*

As can be seen clearly in the deviations, we chose to alter the original plan. The reason for this is threefold.

- Firstly, we are of yet not in possession of the necessary setup containing laser-diagnostics. This makes it unable for us to make a distinction between thermal and radical conversion, nor are we able of measuring the temperature inside and close to the plasma.
- Secondly, we started by downstream injection of argon with methane, instead of synthetic barn air. We expected that usage of the first would make the reactions and following analysis a lot easier.
- Thirdly, we tested the plasma forming gas in the following order: H_2, N_2, Air, and O_2. This is a change from the original plan, that we made due to safety challenges with pure O_2.

1.1. SAFETY MEASURES

As oxygen is a highly reactive substance, the necessary precautions needed to be taken. The entire setup is already placed inside of a faraday-cage, which is a standard safety precaution for reactors using microwaves. The faraday-cage is also strongly ventilated, which is a standard safety precaution for reactors using gasses. The following extra measures have been taken specifically for our own experiments:

- Due to the usage of oxygen, we have chosen to surround the setup by oxygen-sensors, which notify us if the measured percentage of oxygen becomes too high. If this occurs, this would notify us of any leakages, meaning that we need to take action.
- Quick simulations show us that during our experiments with oxygen, nitrogen and methane, there are parameters for which we could end up making NO, NO_2, CO or HCN, all of which are substances that are considered hazardous in excess. For these reason, multiple sensors have been placed surrounding the setup.
- As oxygen is a highly reactive substance, it is necessary to only use materials which do not react. For this, specific O-rings, pressure membranes and tubing has been bought and used.

1.2. PARAMETER-SPACE

Within our project, we repeatedly mention the used parameters and parameter-space. Most are self-explanatory, but the following could use some extra explanation:

- **Plasma Injection Distance:** The distance between the beginning of the waveguide and the middle of the injection of our plasma forming gas. This distance is kept constant and has been minimized, since previous measurements indicated that the shortest possible distance would lead to more stable plasmas.
- **Downstream Injection Distance:** The distance between the end of the waveguide and the middle of the downstream injection point. This is our chosen parameter to define the downstream-point, as we

are not certain where the actual middle of the plasma is located. Minimum possible distance is 2 cm, maximum 14 cm.

- **Absorbed Power**: The applied power used at the power source of our microwave generator, is not the same as the power that actually powers the plasma. For this reason, we measure both the applied and reflected power, which ends up giving us the power absorbed by the plasma, which is the relevant parameter..
- **Reactor pressure**: The pressure at which our plasma-reactor is kept constant. For the purpose of the CANMILK-project, this pressure should be atmospheric. For ease, we keep it at 1000 mbar ± 2mbar, with one exception: hydrogen.

2. DESCRIPTION OF ACTIVITIES

2.1. SETUP

The setup seen in figure 1 is used within our experiments:

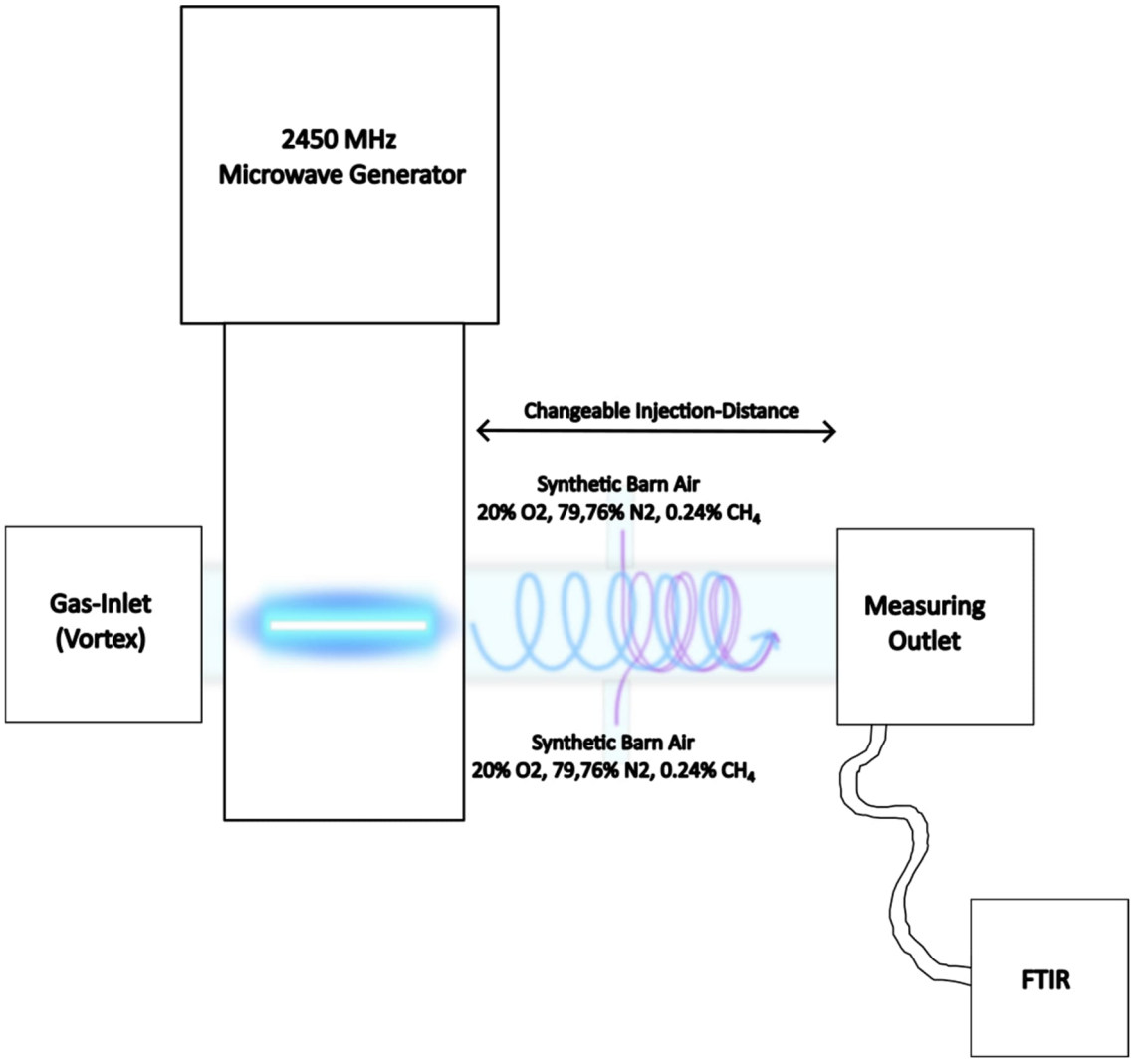

Figure 1: Experimental Setup Drawing

A short explanation about the different components within this setup:

- **Gas-Inlet:** Our gas-inlet consists of two metal gas tubes coming together into a single cylinder. Due to them having a slight offset compared to each other, they are capable of creating a vortex flow (once the flow has become high enough). This vortex flow is necessary to keep the plasma from touching the walls of the quartz tube, preventing it from melting.
- **Microwave Generator:** The solid state microwave generator has a nominal maximum power output of 1 kW at 2.45 GHz. The maximum power output of 1 kW leads to a maximum absorbed power between 710 -730W. Microwaves are applied to the plasma through WR340 waveguides with an automated impedance matching system. The plasma is generated inside a 30 mm OD quartz tube is passed through a WR340 waveguide
- **Changeable Injection-Distance:** We have specific quartz tubes made, which contain two small perpendicular tubes. These can be used as a downstream injection point for our synthetic barn air, where it can mix with our plasma forming flow.
- **FTIR:** In order to do our performance measurements, we need a way to quantify the concentration of species within our reactor. We have chosen to do this via FTIR spectroscopy, as it is a fast, precise and non-destructive method. To analyze the data gained from the FTIR, we have created our own Python-GUI, which uses the python-module RADIS as its core-functionality

2.2. PLASMA-PICTURES

Within the original planning of this project, the idea was to do research into two gasses, which are hydrogen and oxygen. However, in the end we ended up doing research into four different kind of plasmas.

- The **hydrogen plasma** is capable of supplying us with hydrogen-radicals, which can be used to turn CH_4 into C_xH_x. Sustaining the hydrogen plasma is quite difficult. Within our setup, the highest pressure that we could reach was 450 mbar, for which 730W absorbed power was necessary. Hydrogen also needed a higher flow to remain stable, especially at higher pressures. Its most stable condition was found at 42 SLM (*figure 2a, top-left*).
- The **oxygen plasma** is capable of supplying us with oxygen-radicals, which can be used to turn CH_4 into CO, CO_2 and H_2O. As CO is a hazardous substance, it is important that we research the amount that it is created. For this plasma, the lowest stable point at atmospheric pressure was found at 375 W absorbed power and 10 SLM, giving us a wide parameter-space (*figure 2b, top-right*).
- The **nitrogen plasma** is capable of supplying us with nitrogen-radicals, which can be used to turn CH_4 into HCN and H_2. As HCN is an especially hazardous substance, it's important that we research if it's actually created. For this plasma, the lowest stable point was found at atmospheric pressure was 320 W absorbed power at 10 SLM (*figure 2c, bottom-left*).
- The **air plasma** is capable of supplying us with nitrogen- and oxygen-radicals, which can be used to turn CH_4 into HCN, CO, CO_2 and H_2O. As both HCN and CO are hazardous substances, it's important to understand the chemistry happening. For this plasma, the lowest stable point was found at atmospheric pressure was 299 W absorbed power at 10 SLM (*figure 2d, bottom-right*).

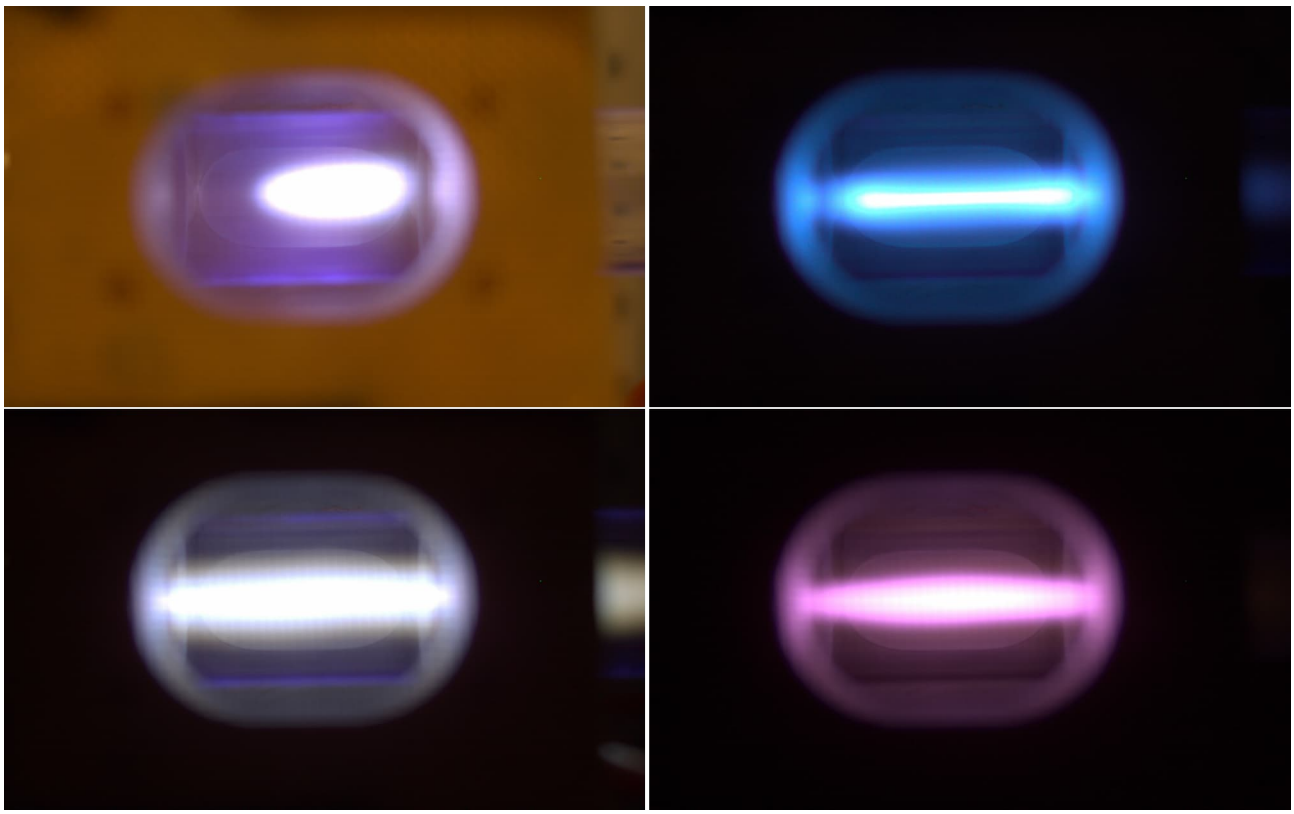

Figure 2: Pictures of the used plasmas (a-d)

2.3. ANALYTICAL FTIR GUI

As shown in our setup, we use a FTIR spectrometer to quantify the concentration of species gained from our reactor. This can be used to calculate how much CH_4 was converted, as well as what species it has been turned into. The most commonly used procedure for quantification would be to take many calibration measurements for the different species, their concentrations, temperature and pressures and use these to calculate the correct concentrations found within our measurements.

However, we have chosen to go the mathematical route, using the python module RADIS. This module makes it possible to calculate the theoretical spectra for different concentrations, temperature and pressures. These theoretical spectra can be used to fit to our experimental spectra and return to us the concentration of the different species. In order to make this easier for ourselves (and for colleagues that will use the FTIR), we have put this into a downloadable .exe python GUI. More information about this GUI and the GUI itself can be found here: https://github.com/StijnHelsl98t/GUI_FTIR

2.4. ARGON-METHANE MIXTURE

As explained within the introduction, we have opted to start with argon-methane mixture instead of synthetic barn air. The percentage of methane within this mixture is 0.1% (1000 ppm).
The decision to use this argon-methane mixture was based on the hypothesis that adding more gasses (N_2, O_2 and CO_2) would make the chemistry and analysis more difficult, so instead we opted to start with what seemed to be the easier option. Our argon-methane mixture actually ended up being used for calibration of our FTIR as well as the creation of our Python-GUI. However, we were also capable of doing two full experiments with this, both described in separate chapters below.

2.4.1. HYDROGEN PLASMA

As we were not capable of starting with an oxygen plasma, we instead opted for the secondary option provided within the original CANMILK plan, which was a hydrogen plasma. Using hydrogen-radicals, we would expect to find the following reactions:

$$yCH_4 + 2H \rightarrow C_2H_x + (2y - \frac{x}{2} + 1)H_2$$

, where the most important possible products are C_2H_2, C_2H_4 or C_2H_6. These products should be visible during the analysis of our experiments, so we should be able to check if these reactions indeed happen.

These experiments were done for the following parameters:

PLASMA-GAS FLOW	INJECTION GAS FLOW	DOWNSTREAM INJECTION DISTANCE	ABSORBED POWER	REACTOR PRESSURE
16-50 SLM	2-12 SLM	2-14 cm	300-730W	50-450 mbar

As one can see, a big parameter space has been considered, but no conversion was found. We hypothesis that this is due to the surplus of hydrogen radicals interacting with the created C_2H_x.

As these results were not expected beforehand, we decided to see if we could validate them via simulations. To achieve this, we employed a 1D plug-flow reactor model using the Cantera Python module. The outcomes are depicted in Figure 3, illustrating the effects of progressively introducing argon into the argon-methane mixture. In figure 3a, we present the overall percentage of the maximum methane as a function of distance within our reactor. As we can see, when we first introduce argon, the conversion actually becomes better. This is as expected, since there is less CH_4 to be converted. However, upon further CH_4 addition, we start seeing that even though the initially injected CH_4 is entirely converted, the overall final conversion declines.

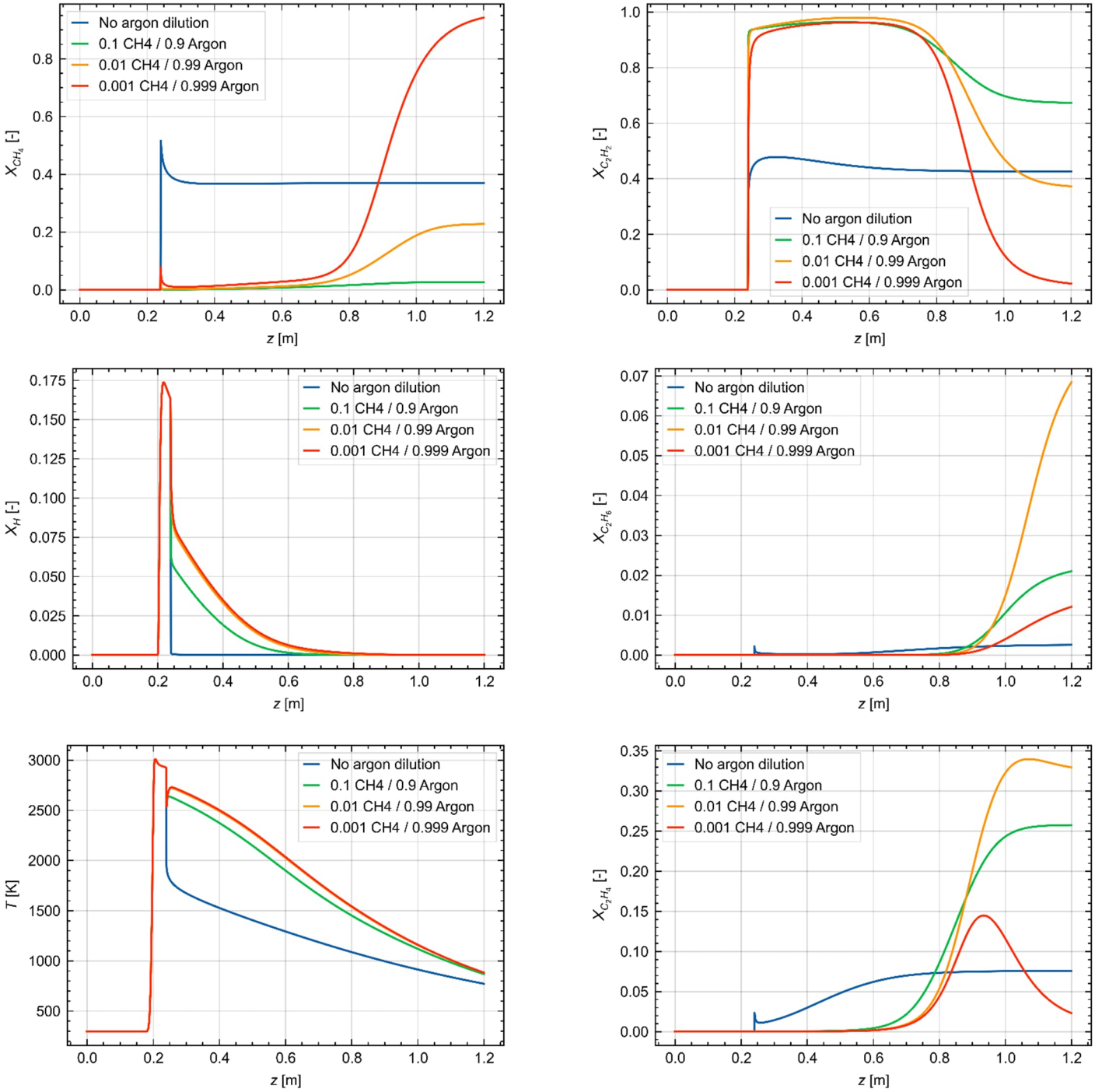

Figure 3: Simulated Data (a-f)

Up to this point, the observed outcomes align with our experimental results. We extend our examination to include the total concentration of hydrogen radicals, as depicted in Figure 1e. Here, the results differ from our hypothesis. For as long as there are still hydrogen-radicals left, the CH_4-concentration remains low. Contrarily, once all the hydrogen-radicals are gone, the methane concentration rises again. This prompts a revised hypothesis that the re-creation of methane happens due to the elevated temperature left within the reactor (shown in figure 1f).

2.4.2. NITROGEN PLASMA

As a trial experiment (partly for another project), we also looked at a nitrogen plasma as an option for conversion. Using nitrogen-radicals, possible reactions could be:

$$2CH4 + 2N \rightarrow 2HCN + 3H_2$$
$$3H_2 + 2N \rightarrow 2NH_3$$
$$2H_2 + N \rightarrow NH_4$$

, where the most important possible products are HCN and NH_x. These products should be visible during the analysis of our experiments, so we should be able to check if these reactions indeed happen.

This experiment was done for the following parameters:

PLASMA-GAS FLOW	INJECTION GAS FLOW	DOWNSTREAM INJECTION DISTANCE	ABSORBED POWER	REACTOR PRESSURE
20 SLM	5 SLM	2 cm	575W	1000 mbar

Within figure 4, one can see the moment the downstream-injection began. On the left, a nitrogen plasma with no downstream-injection is shown.

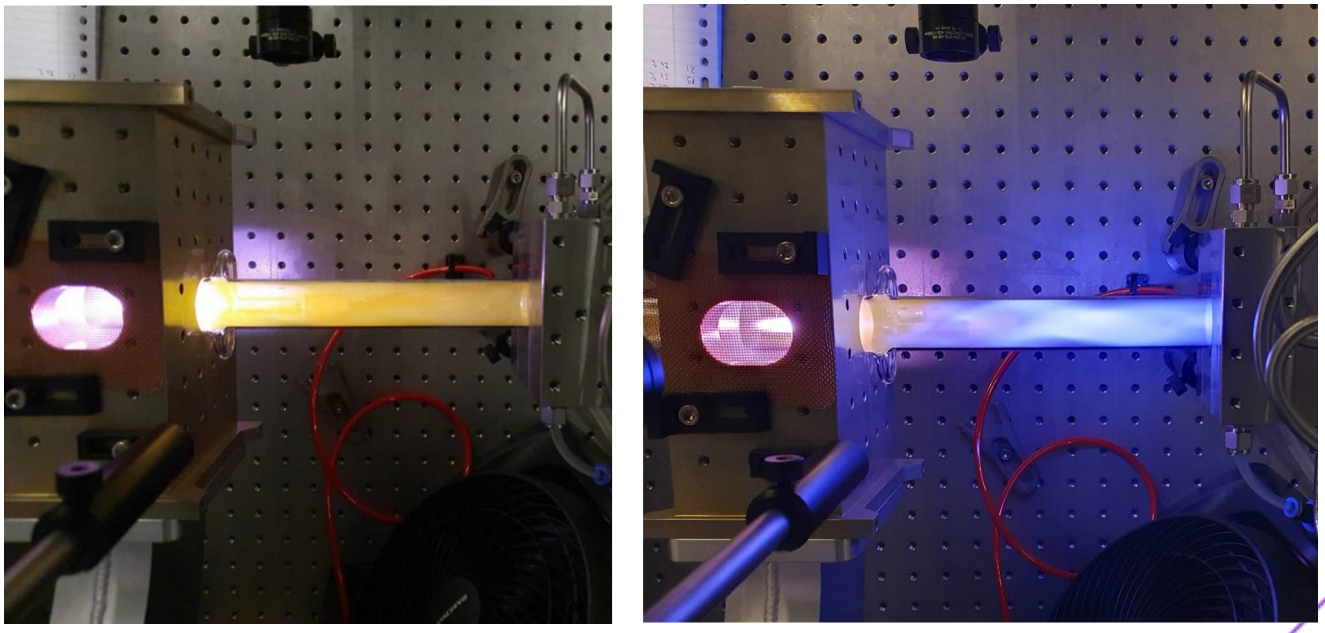

Figure 4: Nitrogen Plasma, HCN creation

On the right, one can see the moment the barn-air is injected. The transition is noticeable, shifting from a vivid yellow glow to a blue hue, attributed to the formation of hydrogen cyanide (HCN). As this substance is highly hazardous, we discontinued the experiments with nitrogen following this event, so no additional spectra were taken.

2.5. SYNTHETHIC BARN AIR

As we do not have possession of actual barn air, instead we opted to make our own synthetic version. For this, we combine O2, N2 and CH_4, resulting in the following composition: 20% O_2, 0.24% CH_4 and 79.76% N_2. This mixture is in our opinion close enough to the actual values within air, but does not include any trace gasses such as CO_2, H_2S, NH_4, etc.

2.5.1. HYDROGEN PLASMA

As we were not capable of starting with an oxygen plasma yet, we instead opted for the secondary option provided within the original CANMILK plan, which was a hydrogen plasma. Using hydrogen-radicals, we would expect to find the following reactions:

$$yCH_4 + 2H \rightarrow C_2H_x + (2y - \frac{x}{2} + 1)H_2$$

, where the most important possible products are C_2H_2, C_2H_4 or C_2H_6. These products should be visible during the analysis of our experiments, so we should be able to check if these reactions indeed happen.

This was done for the following parameters:

PLASMA-GAS FLOW	INJECTION GAS FLOW	DOWNSTREAM INJECTION DISTANCE	ABSORBED POWER	REACTOR PRESSURE
16-50 SLM	2-12 SLM	2-14 cm	300-730W	50-450 mbar

Within this parameter-space, we consistently observed a 100 percent conversion. However, we do not measure the expected CxHx compounds. Instead, we detect H_2O, CO and CO_2. This indicates that instead of thermal or radical conversion, we would have incomplete combustion. Our analysis suggests that the H-radicals convert the oxygen inside of the air, instead of the methane. This reaction is unstoppable, as the activation energy barrier of this reaction is zero. Consequently, a substantial amount of energy is dissipated, as the methane cannot be efficiently destroyed without concurrently converting atmospheric oxygen. An example of a hydrogen plasma measurement is shown in figure 5, which has been done for the following parameters:

PLASMA-GAS FLOW	INJECTION GAS FLOW	DOWNSTREAM INJECTION DISTANCE	ABSORBED POWER	REACTOR PRESSURE
20 SLM	7.5 SLM	2 cm	350-610W	250 mbar

We note the following:

- Firstly, the complete destruction of the injected CH_4 occurs for all tested power levels.
- Secondly, the amount of water we create is still slightly dependent on power, with the maximum value lying slightly above 101000 ppm (10.1%). This aligns with expectations, considering that the oxygen-radicals concentration is approximately ((7.5*20%)/27.5)*2 = 10.9%. This indicates that the majority of oxygen molecules are used to create water, with a minor portion being used to create CO and CO_2.
- Lastly, the total amount of carbon within our reactor seems to be exceed the carbon originating from the CH_4. This suggests the presence of residual carbon soot from prior experiments.

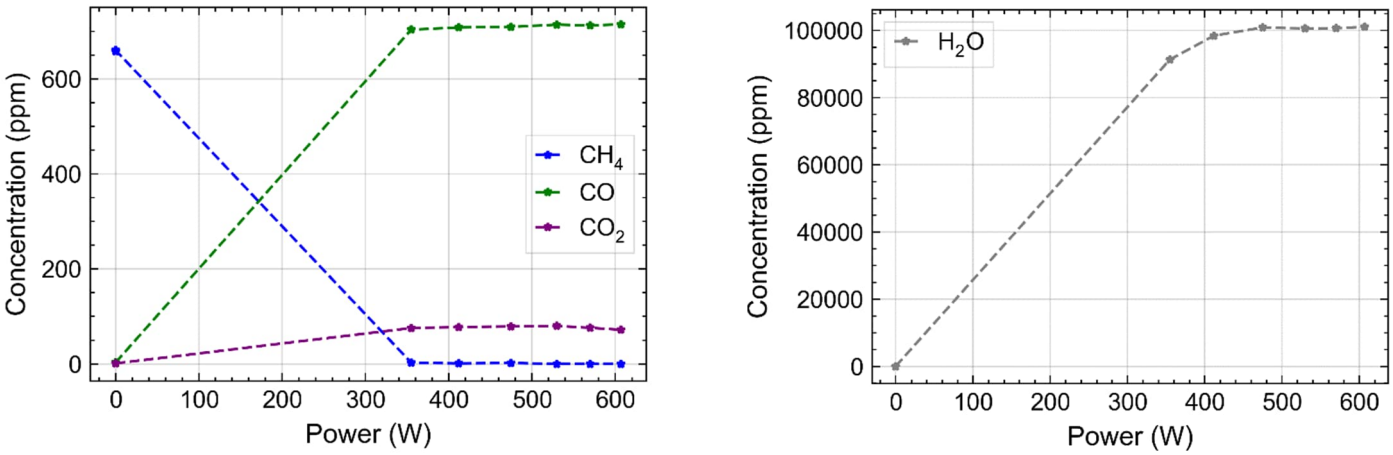

Figure 5: Hydrogen Plasma, Species concentration, single distance experiment

2.5.2. OXYGEN PLASMA

Our second option is oxygen. Using oxygen-radicals, possible reactions could be:

$$CH_4 + 3O \rightarrow CO + 2H_2O$$
$$CH_4 + 4O \rightarrow CO_2 + 2H_2O$$

, where the most important possible products are CO, CO_2 and H_2O. These products should be visible during the analysis of our experiments, so we should be able to check if these reactions occur.

These experiments were done for the following parameters:

PLASMA-GAS FLOW	INJECTION GAS FLOW	DOWNSTREAM INJECTION DISTANCE	ABSORBED POWER	REACTOR PRESSURE
20 SLM	5 SLM	2-14 cm	370-710W	1000 mbar

‘

Within this parameter-space, we are capable of getting good conversion. Within figure 6, shown is the conversion of CH_4 and the formation of new species during an experiment, taken at a downstream injection distance of 6 cm.

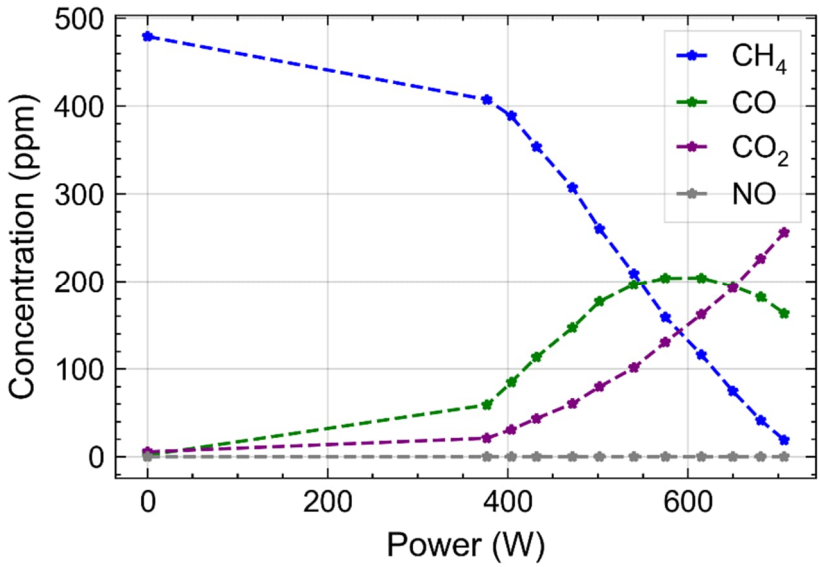

Figure 6: Oxygen Plasma, Species concentration, single distance experiment

Within this figure, the anticipated power curve for CH_4 conversion is clearly evident. From this figure, we find that the CH_4 appears to be converted into both CO and CO_2. However (not shown here), we also notice that the total amount of CO and CO_2 do not always add up. We hypothesize that this missing carbon takes it form as carbon-soot. We also note that there seems to be a power for which we create a maximum amount of CO, beyond which it transitions into CO_2 instead. This finding is noteworthy in itself. Taking thermal equilibrium,

higher power (and thus temperature) should correspond to increased CO production, only decreasing after reaching 4500 K. Though our data does indicate this pattern of increase to decrease, 4500 K seems quite high. Another option could be incomplete combustion, but as our total amount of O_2 is much higher than the amount of CH_4, incomplete combustion seems to be unlikely. Our last possible option would be that radical conversion does play a role

The figure also indicates the absence of NO_x creation. Not shown here are the measured H_2O levels. Despite employing water cooling in our experiments, designed to prevent water contact within the reactor, imperfect seals in the vacuum parts used under one atmosphere conditions could lead to inaccuracies in measured H_2O concentrations, especially at lower values.

Within figure 7, we show the distance dependence of the conversion of CH_4.

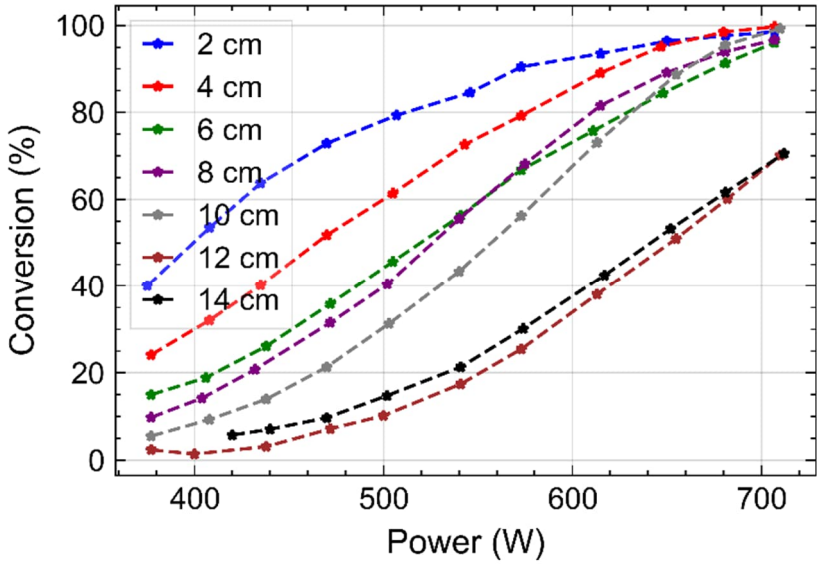

Figure 7: Oxygen Plasma, Distance-dependence of conversion

Two notable observations emerge from this figure:

- Firstly, we note that injecting our barn-air further downstream shifts the conversion-threshold (the point at which conversion starts) to higher powers. This is an expected result, since going further downstream leads to a lower temperature and less oxygen-radicals being present within that point of the afterglow.
- Secondly, we note that despite the conversion threshold is being pushed to higher powers, we are capable of finding improved conversion. This is clearly visible when we compare distance 6 with 8. Even though we have gone further downstream, from 575 W and higher, better conversion is observed at distance 8 compared to distance 6 (the same holds for 10 cm, when we reach 650 W). We hypothesize this is due to mixing playing an important role. As explained, the initial plasma flow forms a vortex. In order for good mixing to happen, our downstream-injected flow needs to penetrate this initial vortex. It appears that at a distance of 6 cm, this initial vortex is still too strong for 5 SLM of downstream-injected flow to reach good penetration. However, at 8 cm, this vortex has grown a little bit weaker, being able to lead to better mixing and thus better conversion, even though the temperature will have gone down and there are less oxygen-radicals present.

We also note that the downstream-injection distance decides the final composition. This can be seen in figure 8. From this figure, the following findings can be concluded:

- Firstly, the NO_x created is highly dependent on distance, where we note that from 4 cm and further, we no longer create NO_x.
- Secondly, we find that the amount of CO created goes up when we go further downstream. Again, this goes against our expectations of thermal conversion. Going further downstream should lead to a lower temperature, again leading to more CO_2 and less CO. Instead, we see a contrary trend. This means again that either the temperature is very high or radical conversion does indeed play a bigger impact.

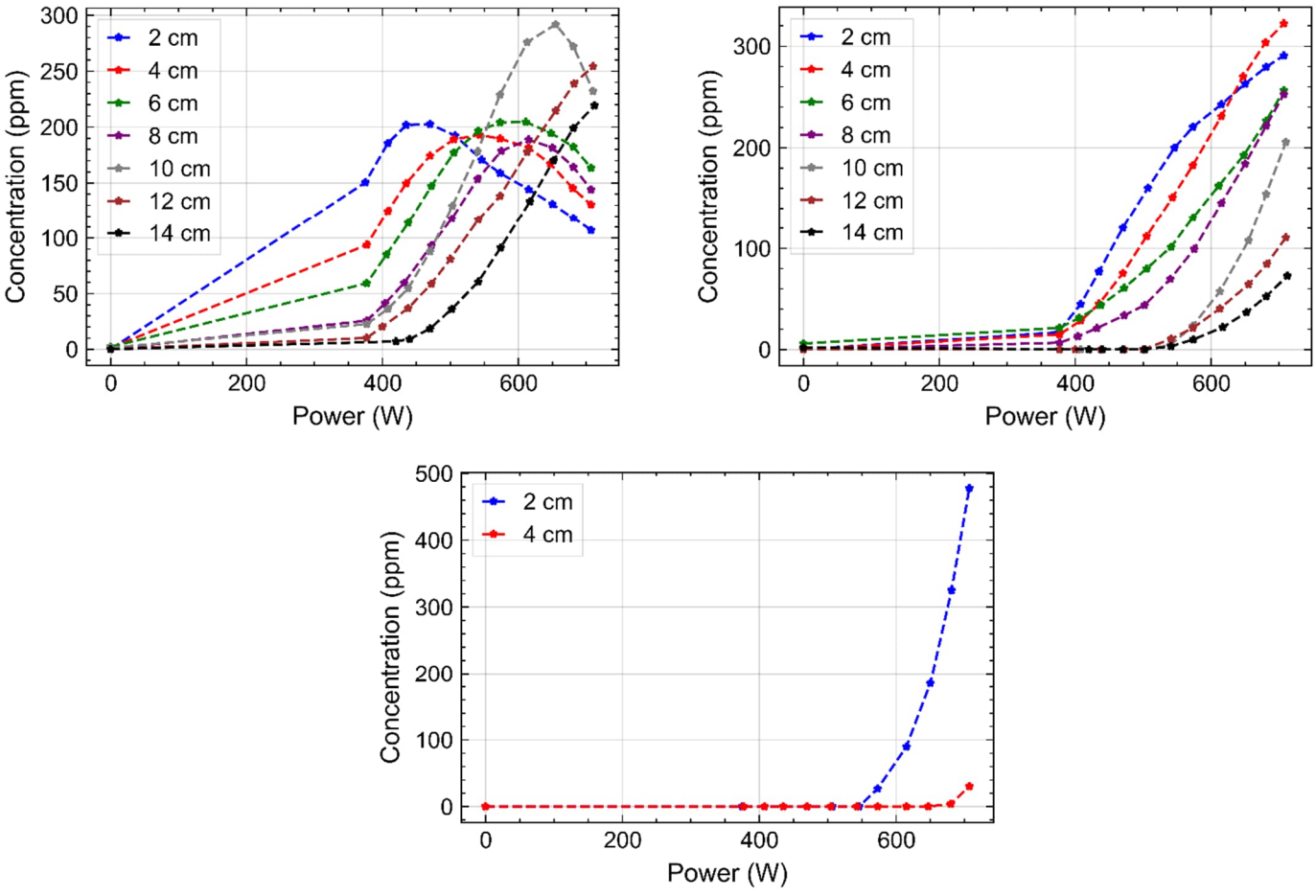

Figure 8: Oxygen Plasma, species concentrations for different downstream-injection distances

2.5.3. AIR PLASMA

As hydrogen may no longer be an option (at least not at one atmosphere, with our power source), we also want to consider an extra option: air (or barn air) plasma. Using an air-plasma, we will be making both nitrogen- and oxygen-radicals. This means that all of the previous reactions mentioned above during the use of a nitrogen- or oxygen-plasma could happen.

These experiments were done for the following parameters:

PLASMA-GAS FLOW	INJECTION GAS FLOW	DOWNSTREAM INJECTION DISTANCE	ABSORBED POWER	REACTOR PRESSURE
20 SLM	5 SLM	2-14 cm	290-710W	1000 mbar

In this experiment, it is crucial to recognize that significant chemistry occurs even in the absence of downstream injection. The presence of both nitrogen and oxygen in the plasma gas results in the formation of NO_x. Additionally, CO_2 is present in the plasma gas, potentially reacting with nitrogen or oxygen radicals. To explore these effects, we compare an air-plasma with no downstream injection to one with downstream injection at a distance of 2 cm. The results are depicted in figures 9 and 10.

From the results, the following observations are noteworthy:

- Firstly, the CO_2 within our plasma-gas is converted into CO, but only a very small amount (max 21 ppm).
- Secondly, injecting barn-air downstream increases the measured NO, but not NO_2.
- Thirdly, when comparing an air-plasma with an oxygen-plasma, it is observed that the total amount of CO created at a downstream injection distance of 2 cm is the same, but with an oxygen-plasma, we need significantly less power to convert this CO into CO_2. This phenomenon could be explained by the fact that in an air-plasma, both energy and oxygen radicals are utilized to generate a substantial amount of NO_x.

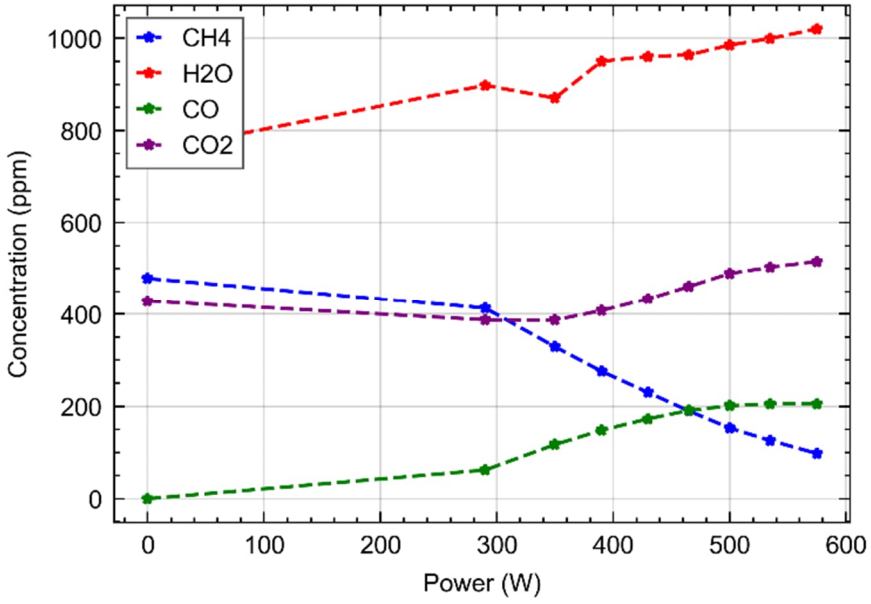

Figure 9: Air Plasma, Species concentration, single distance experiment

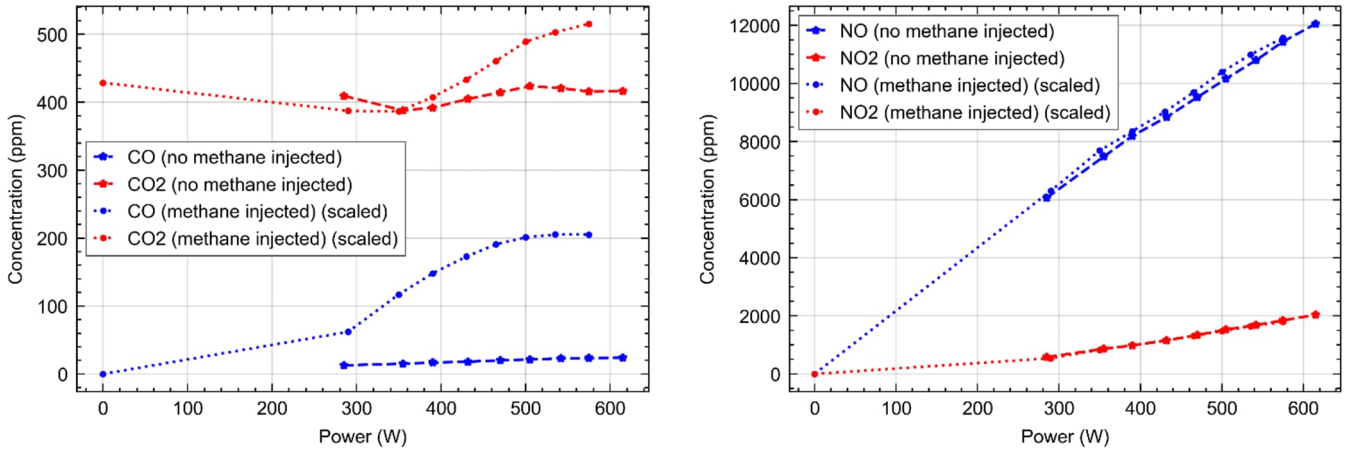

Figure 10: Air Plasma, Comparing no injection with downstream injection

For an air-plasma, we again consider the effects of the downstream injection distance. This effect onto the conversion can be seen in figure 11.

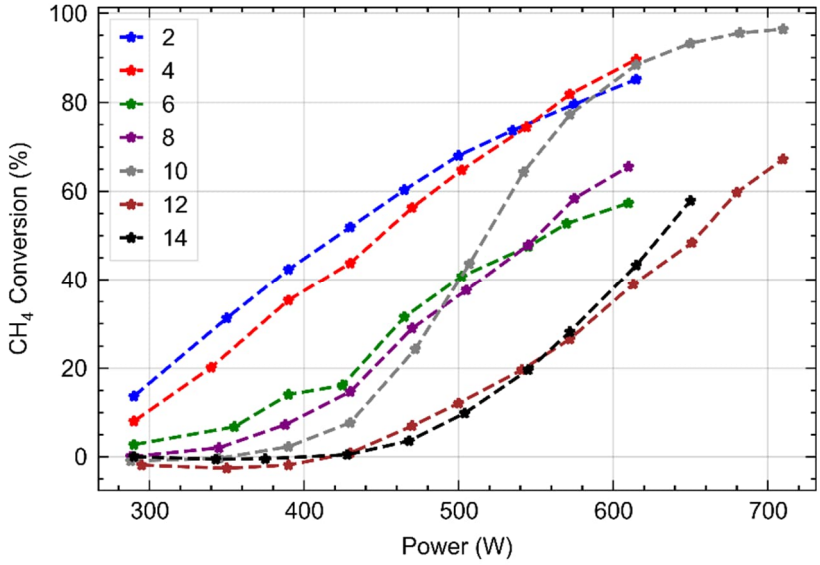

Figure 11: Air Plasma, Distance-dependence of CH_4 conversion

We note that some of the characteristics of CH_4 conversion by an oxygen plasma have remained, whilst others have changed.

Firstly, we note that whilst the same principle remains that at further distances, the conversion threshold is pushed higher powers, we can also see that it's much less clear which distance gives the best conversion. From powers higher than 550, injecting our barn-air 4 cm behind the waveguide seems to give better conversion than at 2 cm. At 6 and 8 cm, the conversion has drastically gone down, while it suddenly peaks again at a distance of 10 cm. We hypothesize that there are two things at play here. Just as we noted previously

with the oxygen plasma, mixing matters. Going further downstream will weaken the vortex of the initial gas, leading to better mixing, which in turn leads to better conversion. However, the difference seems to get larger than previously found with the oxygen plasma. This leads us to our secondary hypothesize, which is that these large differences are due to the amount of NO_x that we are creating.

In an oxygen plasma, the only radicals created are oxygen radicals. These radicals, as well as the thermal energy, are used for only a single purpose: the destruction of methane (see previously, where we show that after 4 cm, no more by-products are created).

In an air-plasma, both nitrogen- and oxygen-radicals are created. Because the plasma will create both radicals, our initial assumption is that the percentage of oxygen-radicals will be lower. As we find no HCN within our final flow, it shows us that the nitrogen-radicals play no part within the destruction of methane. Consequently, only the oxygen-radicals and the thermal energy are used in the destruction of methane. However, the oxygen-radicals and thermal energy can also be used for the creation of NO_x, leading to the reduction of oxygen-radicals and thermal energy being present for the destruction of the methane.

To check our secondary hypothesize, we examine the total NO_x created, as shown in figure 12. Here it becomes evident that (apart from distance 8, where it seems that something might have gone wrong) that the NO_x production does indeed change depending on the downstream-injection distance. As of this moment, we do not see a clear connection between downstream-injection distance and total NO_x created.

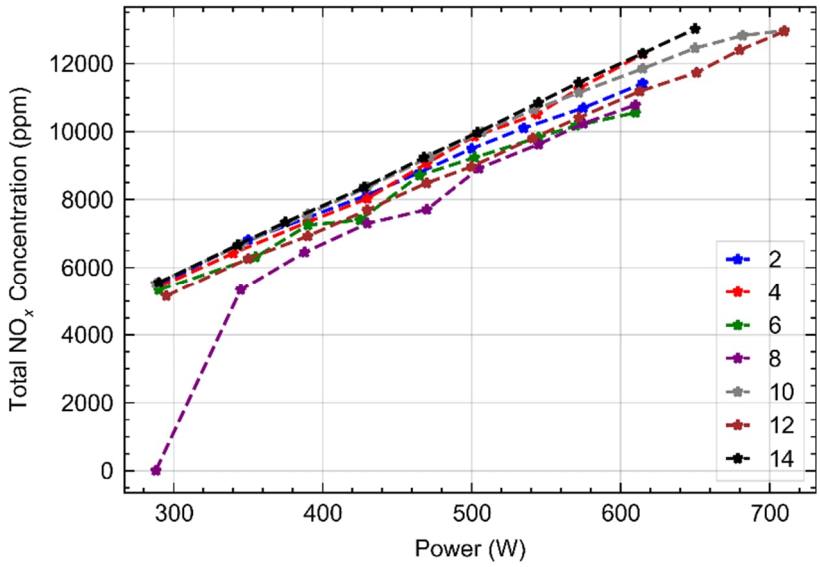

Figure 12: Air-Plasma, NOx production

Lastly, we once again observe that the downstream-injection distance decides the final gas composition, which can be seen in figure 13.

From these figures, we can learn the following:

- The CO concentration becomes higher when the downstream-injection distance becomes higher as well, peaking (within our experiment) at 10 cm, mirroring the behaviour observed in the case of the oxygen plasma.
- The CO_2 concentration becomes lower when the downstream-injection distance becomes higher, consistent with our findings in the oxygen plasma experiments.
- The initial CO_2 from our plasma-gas seems to be destroyed at a distance of 10 cm behind the plasma, not matching any of the other measurements taken. As this matches with our previous finding that at a distance of 10 cm the conversion of methane exceeds the conversion of methane at other distances, we believe this to be a physical finding and not a mistake within the experiment.
- The NO concentration seems to change quite a bit in between experiment, though it does seem to abide to a clear connection with the downstream-injection distance.
- The NO_2 concentration increases with higher downstream-injection distance, peaking at a distance of 4 cm.

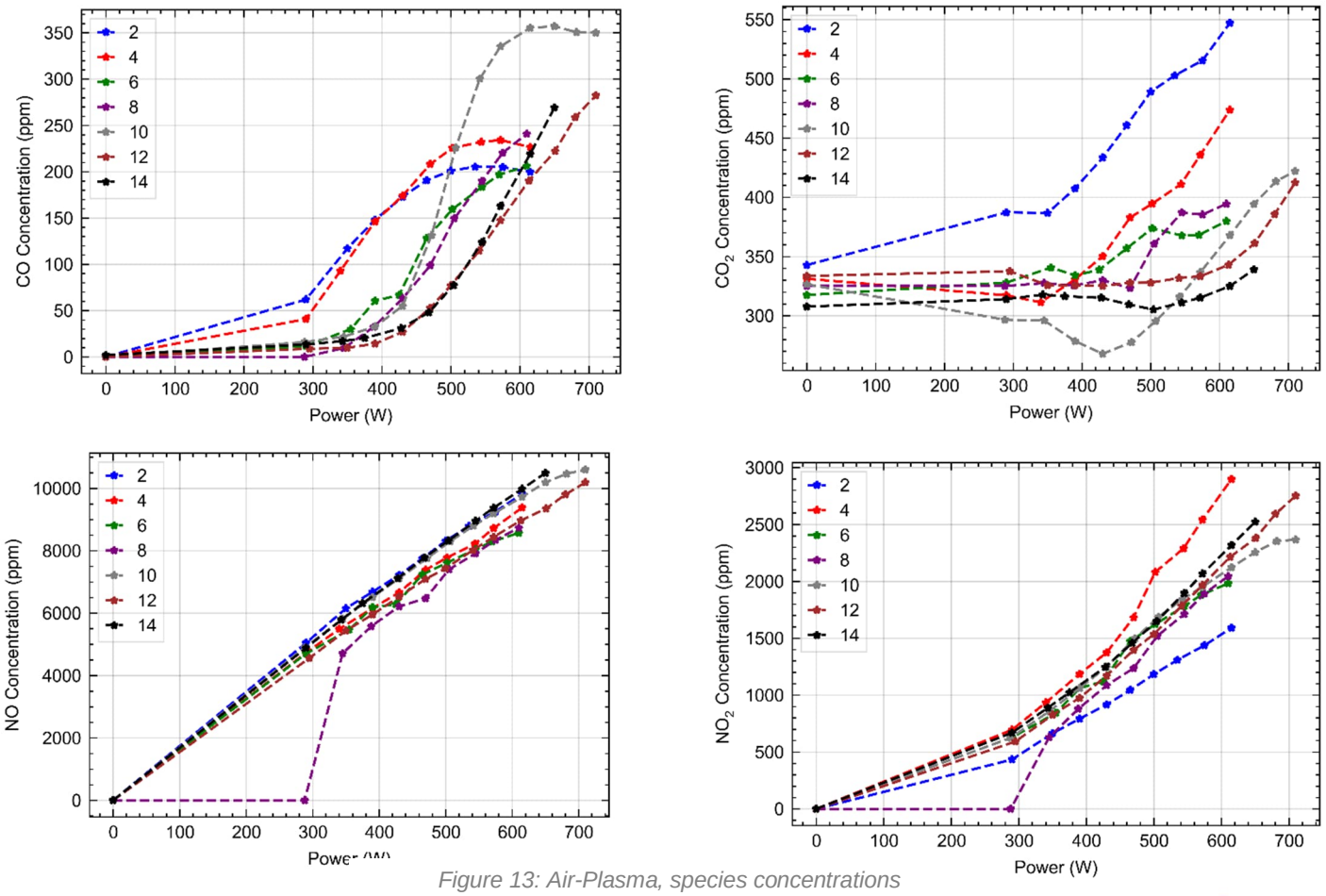

Figure 13: Air-Plasma, species concentrations

3. CONCLUSION

After the first 18 months of this research project, we have been able to build a working setup and started looking into the performance of multiple plasmas. Our main conclusions can be listed as followed:

- The nitrogen plasma creates HCN, rendering it unsuitable for the CANMILK project.
- The hydrogen plasma converts all CH_4 within the barn-air. However, this occurs not through thermal or radical conversion but via incomplete combustion. The majority of the oxygen within the air is used to create H_2O instead. Since the activation energy of this reaction is zero, this cannot be stopped, meaning that we hypothesize that this will never be an efficient way of destroying the CH_4 in the barn-air.
- The oxygen plasma is capable of converting all CH_4 within the air. Depending on the used downstream-injection distance, it will make no NO_x, meaning that this plasma (so far) is a viable option for the original CANMILK plan. However, the downside of using oxygen is its cost and safety concerns.
- The air plasma effectively transforms all the CH_4 within the air, albeit with the additional drawback of generating a substantial quantity of NOx. In order to make this plasma a viable option, one might have to consider finding a purpose for the created NO_x. One purpose could be fertilizer, meaning that farmers could make their own fertilizer whilst cleaning the air from CH_4.

Having primarily conducted performance measurements to date, the next phase of the CANMILK project will focus on preparing the experimental setup for laser diagnostics (leading to a slight deviation from the original WP2.5) and developing the secondary setup for transmission to our partners at VTT.