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**Experimental Investigation of
Plasma Assisted Reforming of Propane**

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Abstract

Plasmatron fuel converters have been investigated for the conversion of a wide range of fuels. This paper is one of a series of papers investigating the performance of plasmatron reformers. In this paper a systematic experimental investigation is carried out of the reformation of propane in a plasmatron fuel reformer. The effect upon reformation of flow rate distribution and electrical power level are investigated for a given geometry that has been thoroughly studied with a similar gaseous fuel, methane. A limited set of startup experiments with propane are discussed

1. Introduction

In a series of accompanying papers [1-5] the performance of a plasmatron methane reformer has been discussed. There is interest on the reforming of methane, primarily driven by methane gas upgrading and conversion to liquid fuels, in order to allow economical utilization of stranded natural gas resources. In addition, that work also provides increased understanding of the behavior of the plasmatron reformer operating with heavier hydrocarbons.

Methane is a gaseous fuel, and its attractiveness in this work has been that we can explore a wide range of parameters without having to be concerned with fuel atomization, wall wetting, and other phenomena that occurs when using liquid fuels. In addition, very complete chemical mechanisms exist that allow adequate modeling of the chemistry. Methane has then been used in the accompanying papers [1-5].

One of the disadvantages of using methane as a surrogate hydrocarbon for heavier fuels is that the chemistry is substantially different. In order to address the issue of the different chemistry, a heavier gaseous hydrocarbon needed to be tested. In this paper, the performance of plasmatron propane reformers is investigated. Propane-air mixture is the simplest hydrocarbon system that exhibits chemical behavior, laminar flame speeds and thicknesses, and extinction limits that are comparable to those of heavier fuels [6]. It is probably the smallest system from which quantitative information directly relevant to the partial oxidation of heavier liquid gasoline and diesel fuels can be extracted, and is therefore an appropriate choice for this study [7]. Unfortunately, unlike the case of methane where chemical modeling of the experiments were performed [3-5], propane mechanisms are not as well developed, and we have not performed chemical modeling to compare with these experiments,

The experimental setup, which is similar to that with the methane experiments, is briefly described in section 2. Results for different methods of injection and air/propane mixing are described in sections 3-7. The results are discussed in section 8. Finally, section 9 gives the conclusions.

2. Description of the setup

The setup used in the experiments, as well as the experimental results with methane, have been reported elsewhere, and will only briefly be described here.

The plasmatron used in the experiments is shown in Figure 1. Three different gaseous flows can be introduced through the wall gas, the swirl (plasma) gas, and the axially injected gas. The wall gas is injected in the axial direction at an axial location similar to the axial nozzle, with no swirl. The swirl (plasma) gas is injected downstream from the wall air and the axial gas, with a large amount of vorticity. The swirl gas moves the discharge into the fuel region, and provides the rotation motion that moves the arc roots on the electrodes (to minimize electrode erosion).

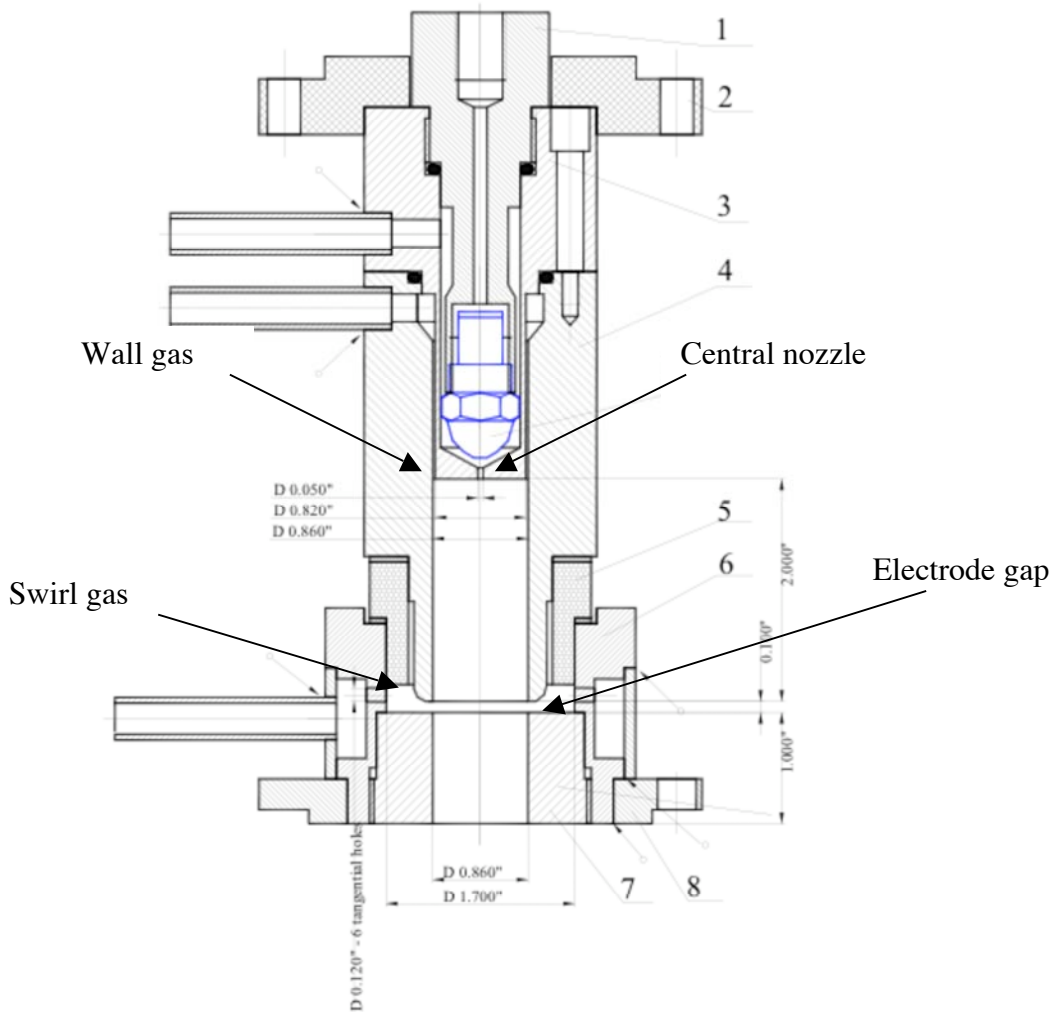


Figure 1. Plasmatron fuel reformer used for experiments with propane.

The experimental results were carried out with propane introduced in 5 different ways: (a) through the axial nozzle, similarly to what is done with liquid fuels (described in section 3); (b) premixed with the wall air (described in section 4); (c) premixed with the swirl (plasma) air (described in section 5); (d) premixed with all the air, so that the air/propane distribution in the plasmatron and the reactor are homogeneous (described in section 6); and finally, (e) stoichiometric conditions in the swirl gas, with the rest of the propane introduced with the wall air (described in section 7).

Downstream from the plasmatron head there is an insulated 12 cm (5 in) long reaction extension cylinder which is also included in the model. The volume of this section is approximately 1000 cm³.

The gas analysis has been described in accompanying papers [1,2]. It consists of a dual column GC and a mass spectrometer. The GC is used for steady state evaluation, and the

mass spectrometer is used for the transient experiments. Soot is measured using a Wagner 2000 opacity meter, but during these experiments the soot level was below detection (0.0% opacity).

2.1 Air/propane mixtures

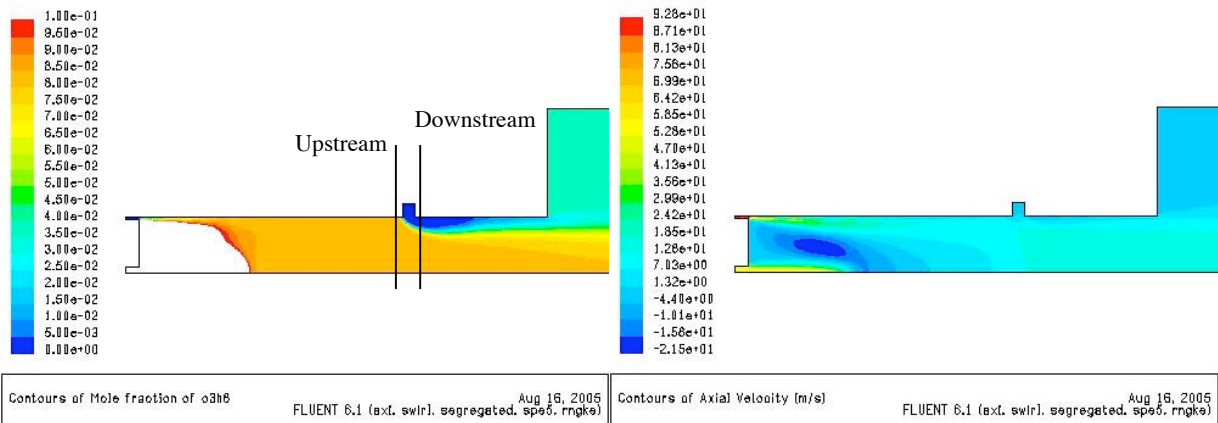
The axisymmetric steady state solution of the flows in a plasmatron propane converter is calculated using a commercially available CFD code, FLUENT. Description of the model and results for the case of methane are presented in an accompanying paper [5]. The model was used to determine the effect of initial conditions on the mixing upstream of the region where sufficient thermal energy has been released to modify the result, which occurs downstream from the plasma region.

Because of the multitude of cases, only three cases will be presented: the case of axial injection of propane, propane premixed with the wall air, and propane premixed with the swirl air. The flow rates used in the simulation of the different injection modes are shown in Table 1. The flow rates for the different modes correspond to those that experimentally optimize the reforming. The flow rate of propane is held constant in the experiments and in the model (at 0.5 g/s), but the total amount of air and the distribution of the air is varied to optimize the reforming.

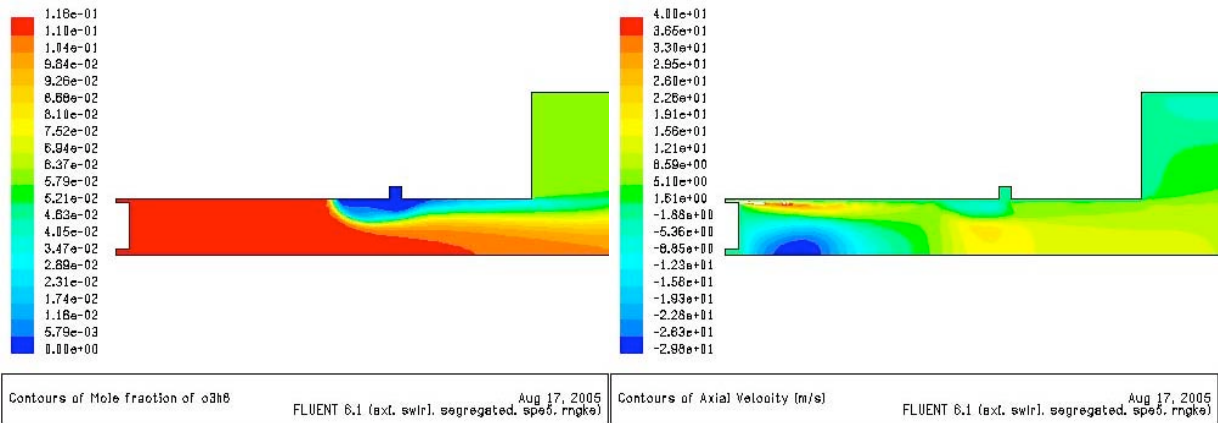
Table 1. Flow rates (in g/s) for the different modes of injection

Fuel injection	Axial	Premixed wall	Premixed plasma
plasma air	1.29	1.51	1.51
plasma fuel	0	0	0.5
wall air	3.67	2.59	1.51
wall fuel	0	0.5	0
axial fuel	0.5	0	0

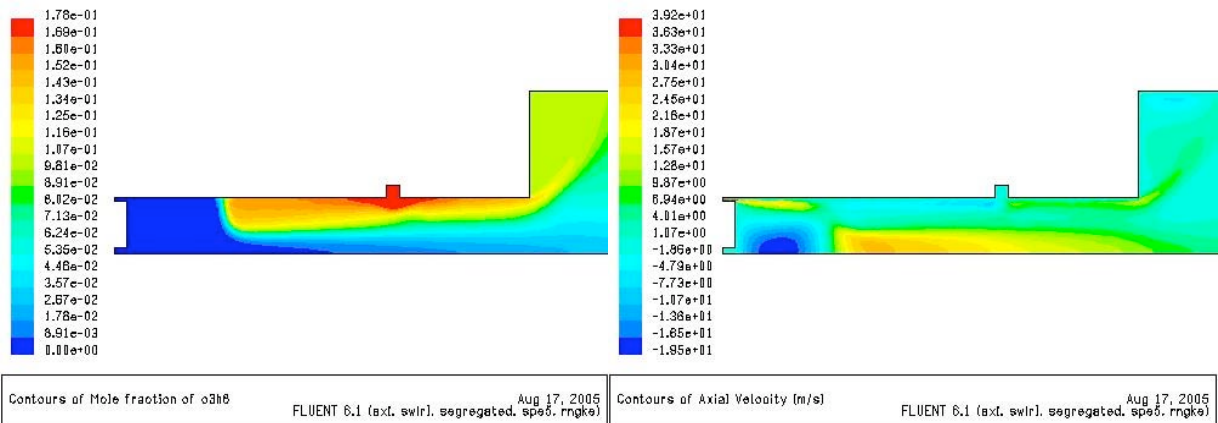
The model indicates very different distribution of the air/propane mixture for the different modes of injection. The propane concentrations for the three modes of injection are shown in Figure 2. Also shown are the axial velocities of the gas for the three cases. The figure shows a cross-section of the plasmatron. The axis is the horizontal at the bottom of the figure, with the radius in the vertical direction, the axial and wall gas injection are at the left of the figure, the swirl gas is introduced in the upper boundary towards the middle of the narrow section, and wide section at the right is the beginning of the reactor. Only a section of the reactor is shown in the figure, as the model is not appropriate for this region because of the absence of chemistry. The figures therefore show half of the plasmatron section shown in Figure 1, rotated counterclockwise by 90 degrees.



(a) Axial injection of propane: molar concentration (left) and axial velocity (right)



(b) Premixed propane with wall air: molar concentration (left) and axial velocity (right)



(c) Premixed propane with plasma air: concentration (left) and axial velocity (right)

Figure 2. Results from CFD calculations of propane molar concentration (left) and axial velocity (right) for the three modes of injection shown in Table 1.

The swirl gas flows upstream from the injection point, because of the strong vorticity. How far upstream is determined by the swirl flow and the axial flows. The case where the propane is premixed with the swirl air results in the largest upstream flow, followed, by the propane premixed with the wall air and finally by the axial fuel injection. The case with the propane premixed with the swirl flow has the lowest wall air flow, followed by the propane premixed with the wall air and by the axial propane injection. This back flow has a large impact on the air/propane distribution at the location of the plasma (around the region of the electrode gas, where the swirl gas is introduced).

In the three cases considered there is a large amount of recirculation upstream the electrodes. In some alternative designs of the plasmatron there is a diaphragm in the region between the axial nozzle and the electrode gap, but it is not needed for the design shown in Figure 1, as the recirculation flow happens automatically. This recirculation has a large impact on the air fuel distribution. For the case of axial injection of the propane, the recirculation results in a very high level of mixing between the propane and the wall air, well before the region of injection of the swirl gas. There is nonuniform air/propane distribution only in the small region between the central zone of the wall air plus propane and the periphery where the swirl air is located. Figure 3 shows the radial molar concentration of propane for a plane that corresponds to the upper electrode and for a plane corresponding to the lower electrode (downstream). The axial locations of these planes are shown in Figure 2 (a, left). The propane concentration is uniform upstream of the electrode gap, and downstream from the electrode gap there are two clearly demarked zones, one with about 8% propane and the second with no propane. For stoichiometric propane air combustion, the concentration of propane is about 4% molar. Thus there is a very small region with easily ignitable fuel.

In contrast, for the case of propane premixed with the swirl air there is a larger zone of stoichiometric air/fuel mixture, with gradients in the opposite direction. The propane molar concentration in Figure 2 b shows larger regions of stoichiometric air/propane mixtures, which should be favorable for two stage reforming, as discussed in the accompanying methane papers [1-5]. Figure 3 shows that indeed there are larger regions of stoichiometric air/propane zone in this case.

The case of propane premixed with the wall air falls between the case with propane axial injection and propane premixed with the swirl air. It will be shown in the experimental section of this paper that indeed, best reforming occurs with propane premixed with the swirl air, followed by premixed with wall air and finally, axially injected propane. The case where propane and air are fully premixed (equal composition of the wall gas and the swirl gas is also discussed in the experimental section. This last case of course has no nonuniform distribution, and the reforming results are comparable to that of the axial propane injection.

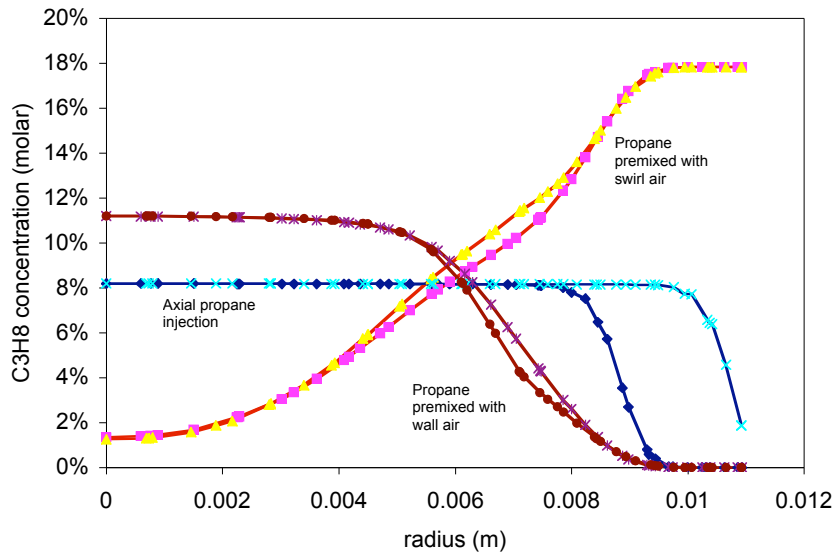


Figure 3. Propane concentration as a function of radius for locations upstream and downstream from the electrode gas, for the three difference modes of injection of propane.

3. Propane injection through axial nozzle

In this section the results of experiments with propane injected through the axial nozzle are presented. As described in the previous section, this configuration results in highly homogeneous air/fuel mixtures, with constant propane in the central region and with large concentration gradients in the region where the air/propane mixtures form stoichiometric combustion mixtures.

The results of the reforming experiments are shown in Figures 4 and 5, as contours of constant quantity as a function of the wall air and swirl (plasma) air, for a propane flow rate of 0.5 g/s. Figure 4 shows the H_2 , CO, CO_2 concentrations and overall O/C ratio, while Figure 5 shows methane concentration, reactor temperature, efficiency and carbon balance. The carbon balance is within 5%. Since water is not measured, it is not possible to close the mass balance for oxygen or hydrogen.

The hydrogen concentration peaks in a band that corresponds to constant O/C ~ 2 . Stoichiometric combustion of propane occurs at O/C ~ 3.33 , and thus O/C ~ 2 is still far away from combustion. Hydrogen concentration is about 12%, with a power efficiency to H_2 , CO, CH_4 and C_2 's of about 60%. At the higher values of O/C more and more of the hydrogen gets combusted, while at the lower values of O/C the conversion of propane is reduced, thus resulting in a maximum concentration as a function of O/C.

The concentration of CO_2 increases with increasing O/C ratio, as expected, as more of the fuel is combusted. The temperature measured by the thermocouple, which is only an indication of the actual gas temperature, increases with O/C.

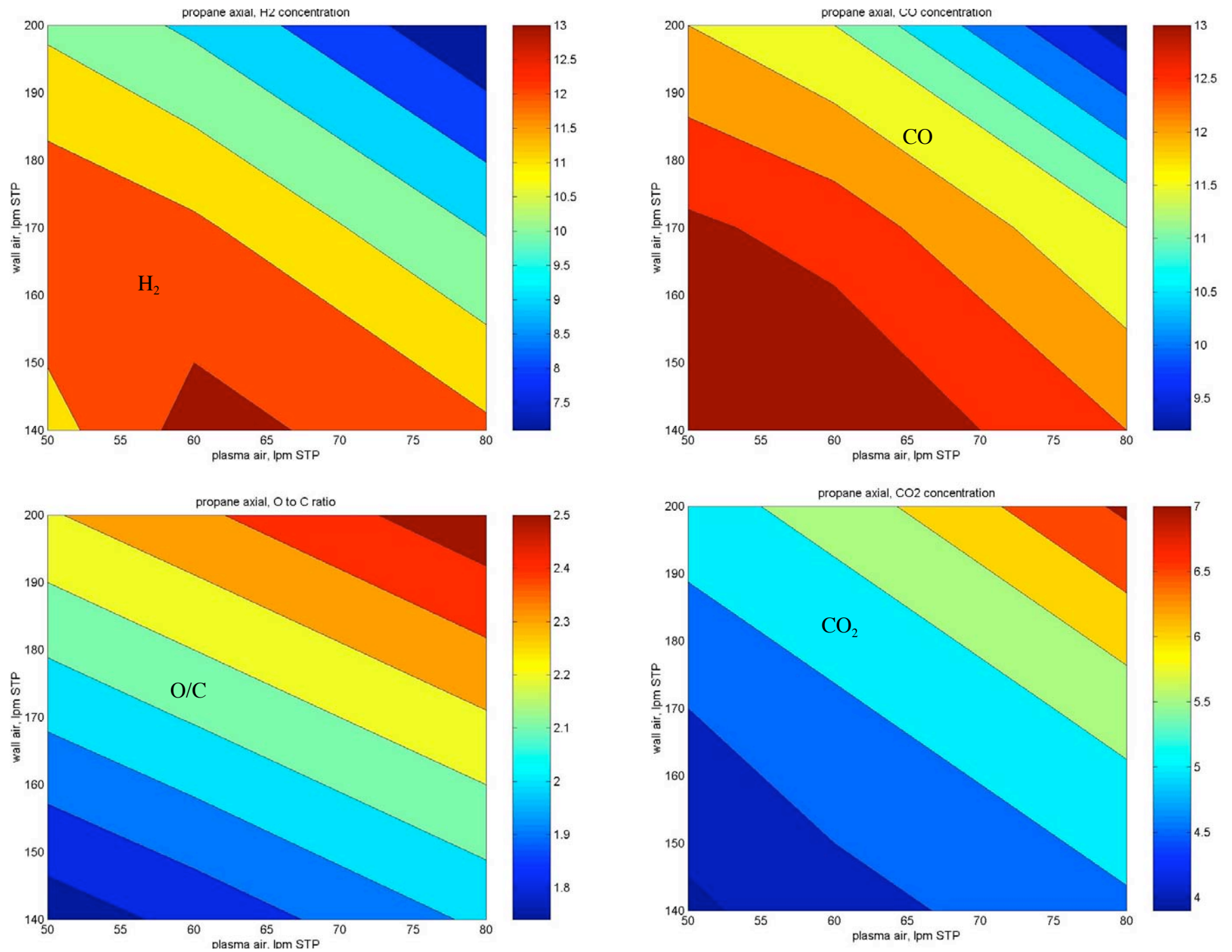


Figure 4. Contours of constant (a) H₂ (b) CO (c) O/C ratio (d) CO₂ as a function of wall and swirl air flow rates, for axial propane injection

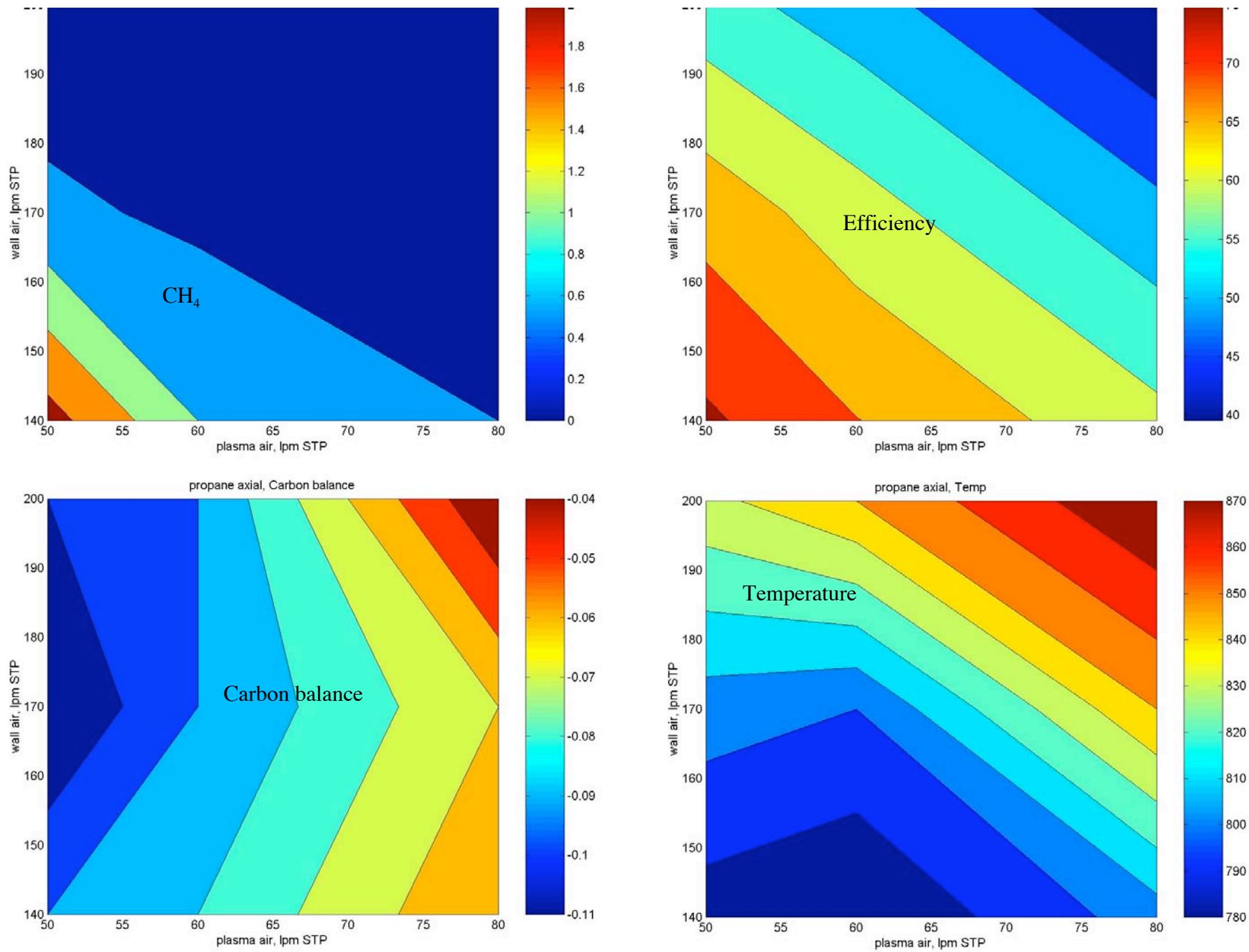


Figure 5. Contours of constant (a) CH₄ (b) efficiency (c) carbon balance (d) temperature as a function of wall and swirl air flow rates for the same conditions as Figure 2.

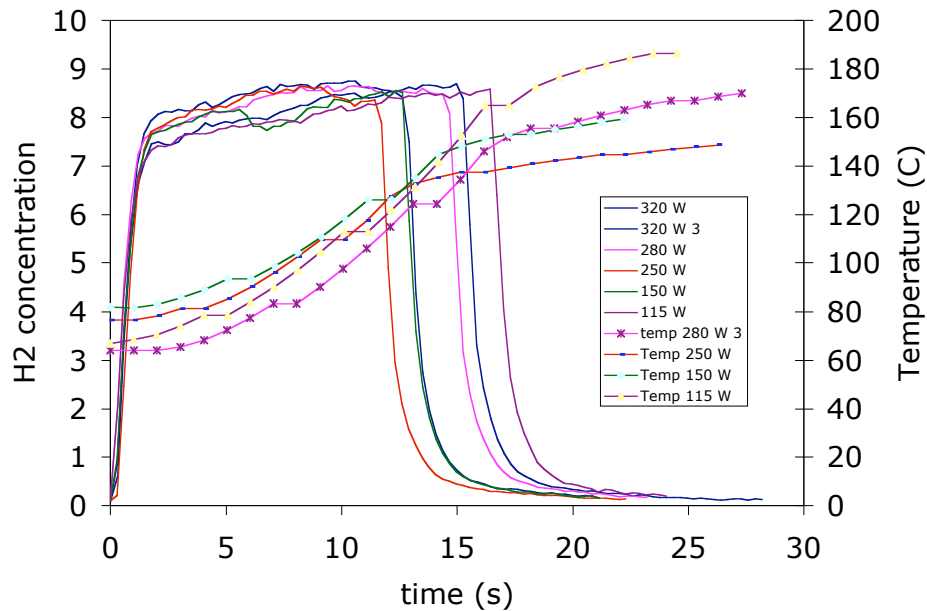


Figure 6. Transient in axially-injected propane for several levels of power, for O/C \sim 2.1

The startup transient characteristics with axially injected propane are shown in Figure 6, for a wall air of 170 lpm and a swirl (plasma) air of 60 lpm. The air/propane flows are established with the reactor at nearly room temperature, and then the plasma is turned on. Figure 6 shows the hydrogen concentration as well as the temperature for different plasma powers. The hydrogen concentration reaches 8% in about 1 s, as was discussed in the transient paper with methane [2]. At this high value of O/C \sim 2.1, the startup is not sensitive to the power level. This was also found to be the case for methane [2]

4. Propane premixed with wall air

Experimental results for propane premixed with the wall air are shown in Figure 7 and 8. The results have similar trends than those in section 3 for the case of axially injected propane. Hydrogen concentration peaks at a given O/C ratio, production of CO₂ increases with O/C ratio. The hydrogen concentration is similar to that of section 4, but occurring at lower O/C ratios, O/C \sim 1.7, vs O/C \sim 2 for axially injected propane. The efficiency to light hydrocarbons is about 55%. As discussed in section 2, the increased nonuniformity of the air/propane distribution is likely to be reason for this result.

The transients are shown in Figure 9, showing 8% H₂ concentration within 1 s, for 120 lpm wall air and 50 lpm swirl (plasma) air, corresponding to an O/C \sim 1.6. The transients are short, about 1 s. In this case the conversion is a stronger function of power than in the case in section 3 that operated at higher O/C ratios. It is also interesting to note that the reformation continued in the absence of plasma. As opposed to the case in Figure 6, where the propane was shut down with the plasma on, in Figure 9 the plasma was shut down with propane still on. This is a feature that has been investigated in the plasmatron startup methane reformer paper [2]. For a power of 120 W, the reforming is poor and the rate of temperature rise is low.

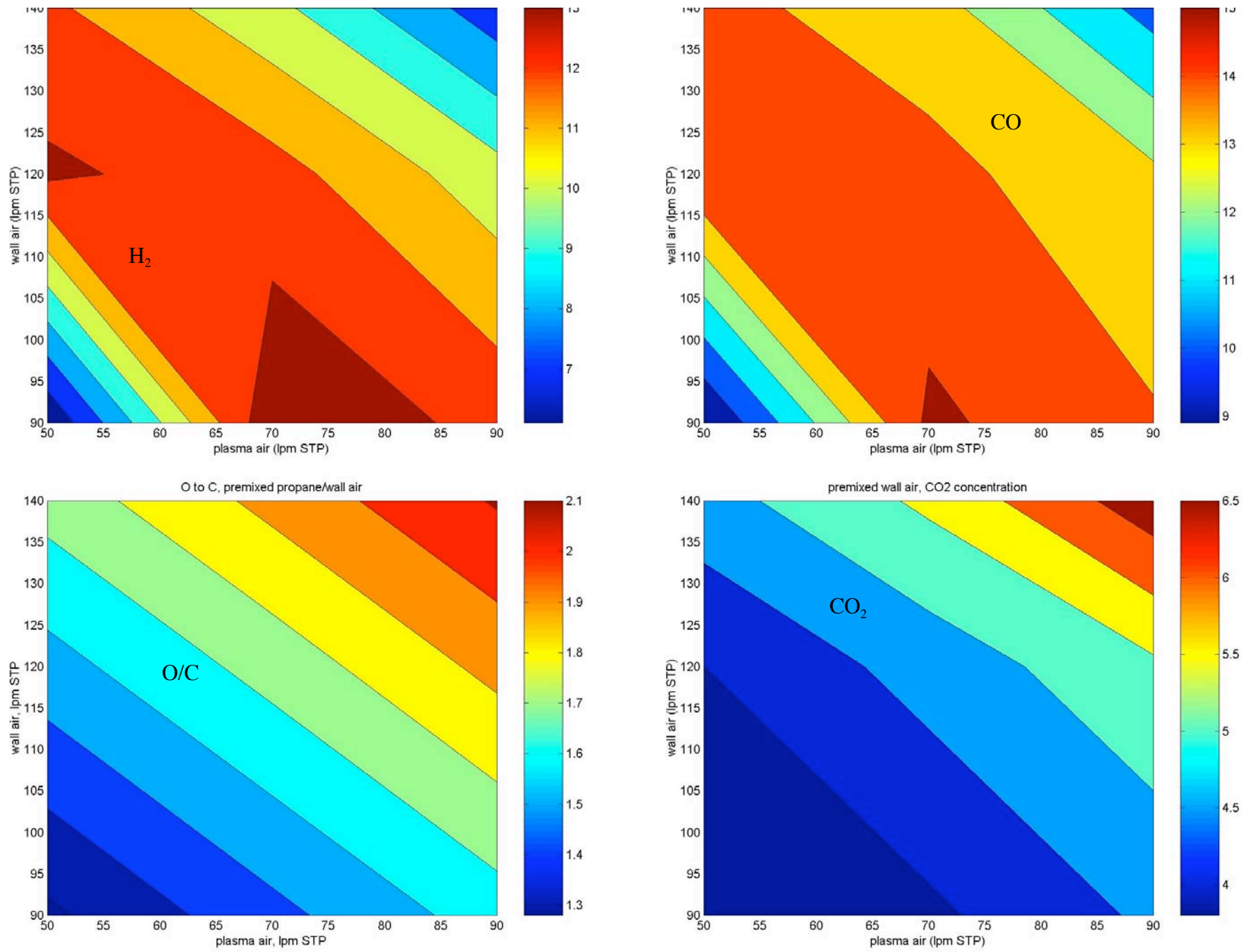


Figure 7. Contours of constant (a) H₂ (b) CO (c) O/C ratio (d) CO₂ as a function of wall and swirl air flow rates, for propane premixed with wall air

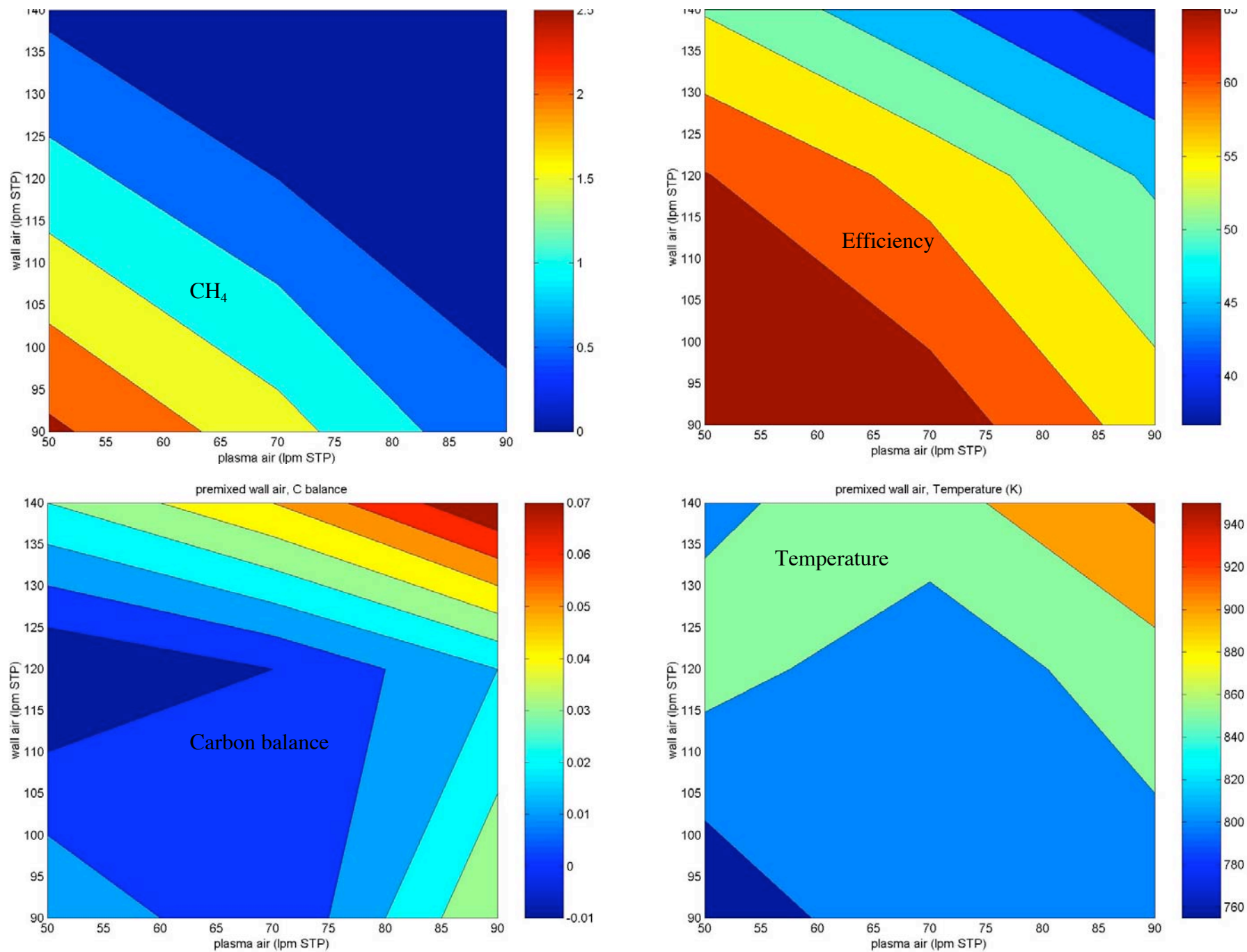


Figure 8. Contours of constant (a) CH₄ (b) efficiency (c) carbon balance (d) temperature as a function of wall and swirl air flow rates for the same conditions as Figure 5.

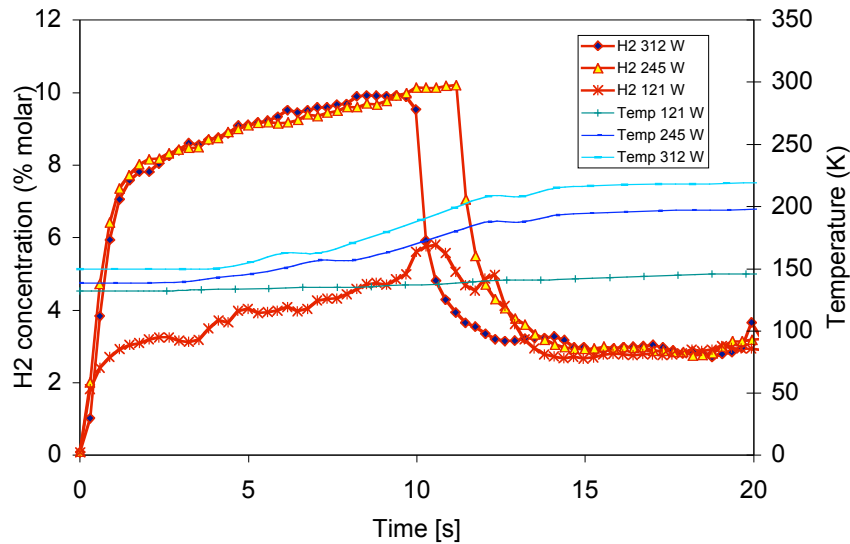


Figure 9. Transient response of plasmatron propane converter operating at $O/C \sim 1.6$, for propane premixed with the wall air, for several power levels.

5. Propane Premixed with plasma air

The fluid dynamic modeling indicated that the air/propane mixtures in the condition when the propane is premixed with the swirl air results in the largest fraction of propane under conditions of stoichiometric combustion (Figure 2(c) and Figure 3). This method of injection was therefore particularly interesting, as the large region with stoichiometric air/propane mixtures should result in faster chemical reaction rates, with the highest release of chemical energy. If the reforming is through the two-stage process, as described in [3-5] (with a fraction of the fuel combusting and distributing the energy through mixing with the rest of the air/fuel mixture), then this case should produce the best reforming results.

The experimental results corresponding to this situation are shown in Figures 10 and 11. The hydrogen concentration in this case is about 14-15%, with 16% CO. The optimal O/C ratio in this case is lower than in the previous 2 cases, $O/C \sim 1.3$. This mode of operation results in substantially lower concentrations of CO_2 , decreased temperatures and increased efficiency ($\sim 80\%$), all pointing towards improved reforming. As a matter of fact these conditions are the best conditions obtained in the study, as will be discussed in section 8. The hydrogen concentration is still substantially lower than that of ideal partial oxidation reforming, in part due to the higher O/C ratio ($O/C = 1$ for ideal partial oxidation). Not only is there additional oxygen, but also the oxygen preferentially oxidizes the hydrogen over the CO. The hydrogen yield is about 45%.

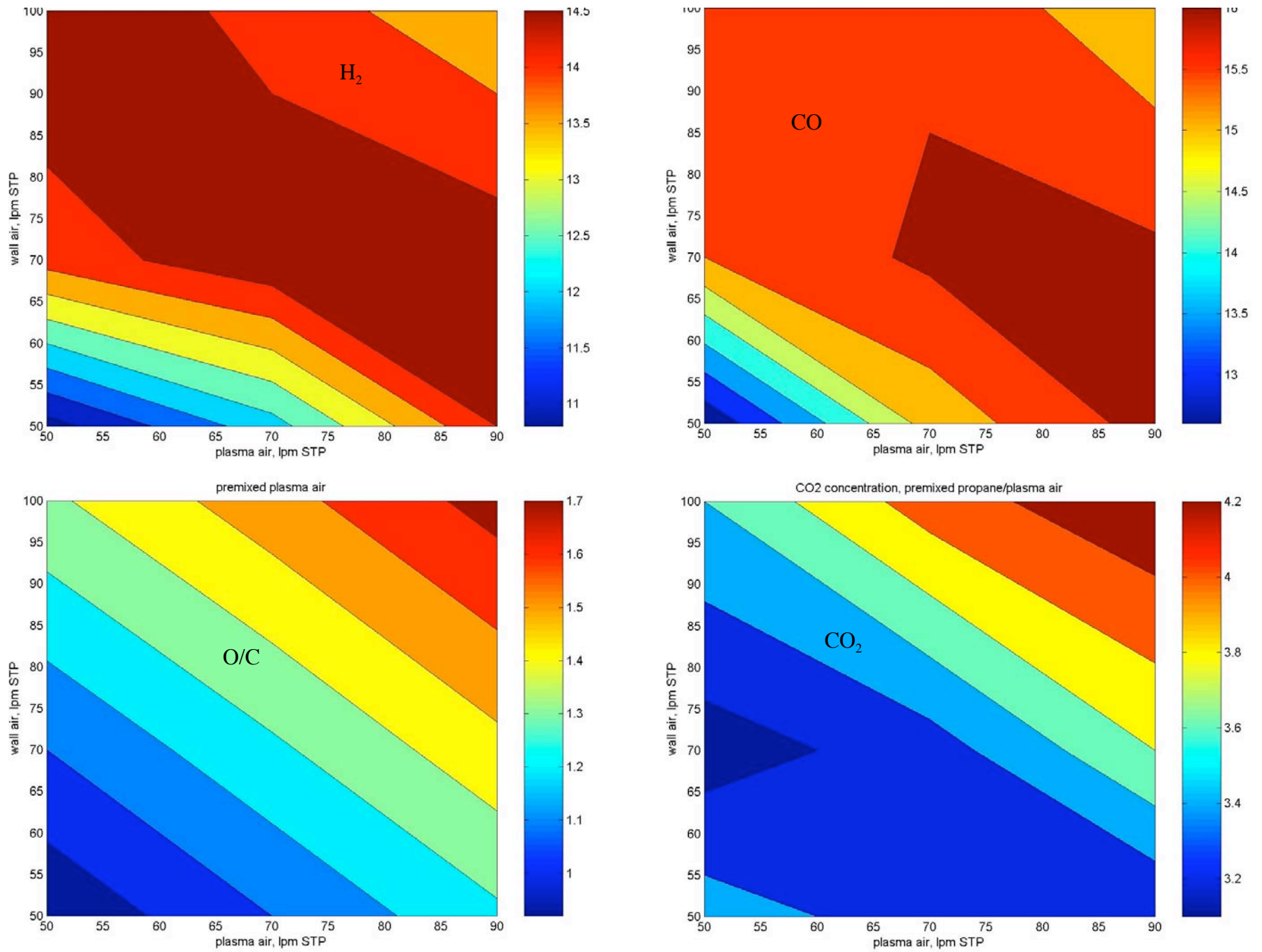


Figure 10. Contours of constant (a) H₂ (b) CO (c) O/C ratio (d) CO₂ as a function of wall and swirl air flow rates, for propane premixed with swirl (plasma) air

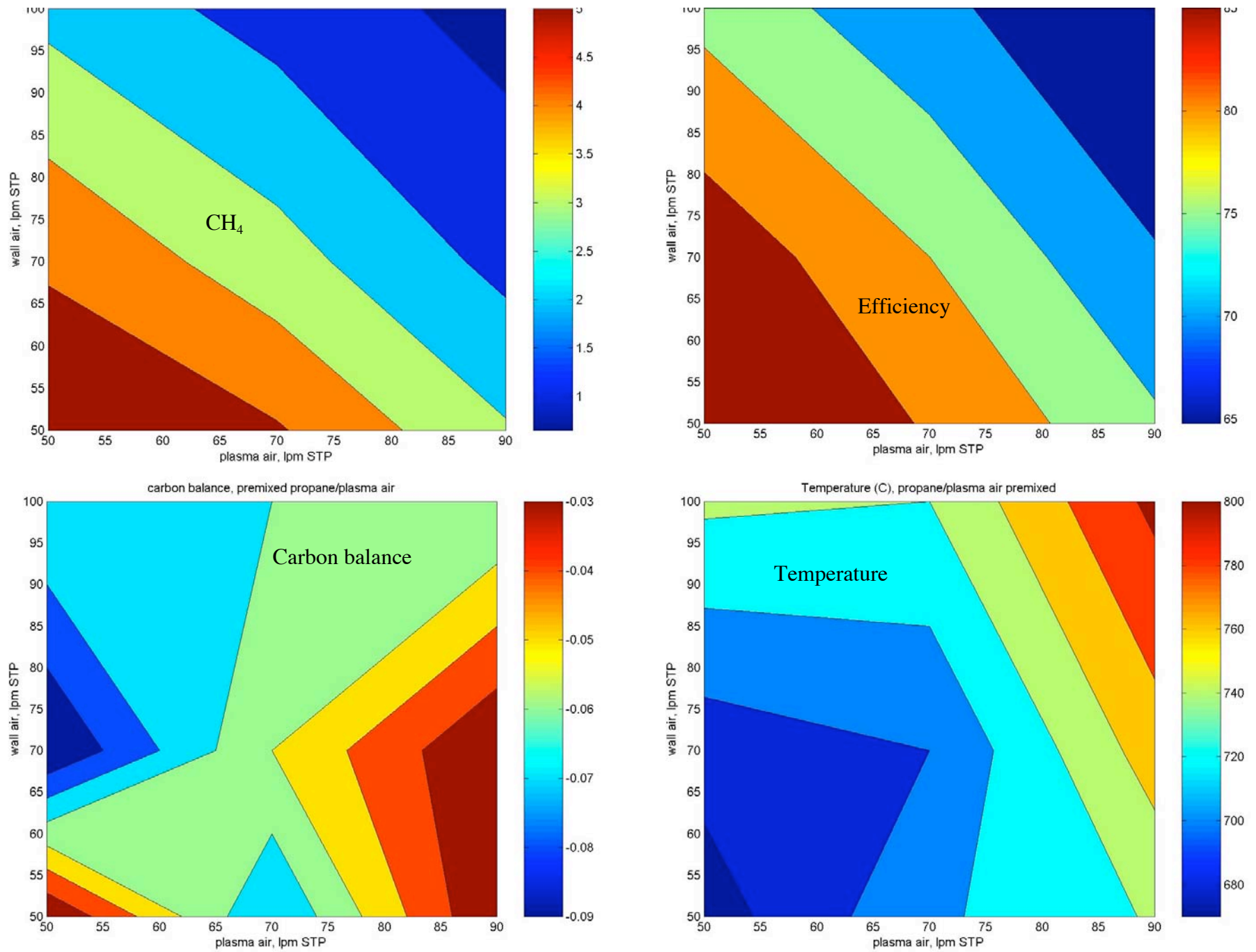


Figure 11. Contours of constant (a) CH₄ (b) efficiency (c) carbon balance (d) temperature as a function of wall and swirl air flow rates for the same conditions as Figure 8.

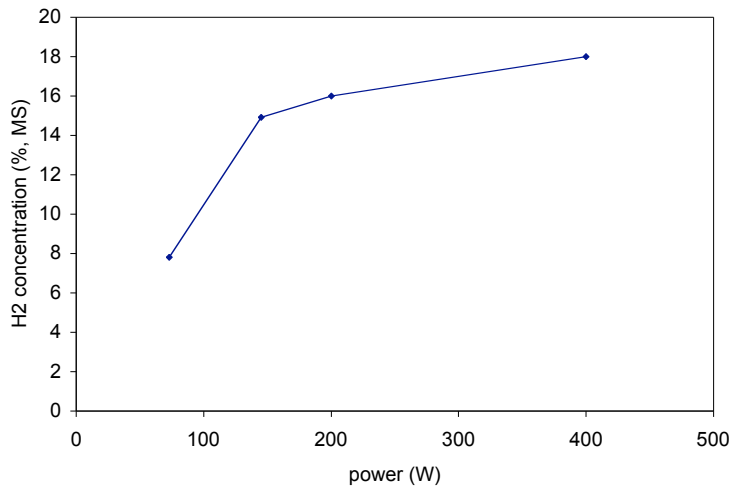


Figure 12 Steady state hydrogen concentration as a function of plasma power (O/C ~ 1.3)

The carbon balance, shown in Figure 11, is better than 10%. The carbon balance is substantially better than this over most of the experimental regime, with the exception of those regions where the reforming is least optimum, at lower swirl/wall air flow rates and where it is likely that there be substantial unconverted propane.

The steady-state power sensitivity is shown in Figure 12, for an O/C of 1.3 (70 lpm for both swirl and wall airs). Higher power does increase the hydrogen concentration.

Transients are shown in Figure 13, for several values of power. The transient hydrogen concentration is a sensitive function of the power, as in the case of for premixed propane/wall air. A hydrogen concentration of 10% can be reached in about 1 s at 400 W. At 100 W, the hydrogen concentration is about 3-4%. Not only is the concentration higher at the higher power, but also the slope after the initial transient is higher. This is due to increased rate of increase of the temperature, not shown in Figure 13. Note that as in the case for premixed propane/wall air, if the plasma is turned off, there is still reforming, although with hydrogen concentrations ~ 2%. The concentration of hydrogen after the plasma has been turned off is not sensitive to the plasma power. Also shown in Figure 13 is one case, one of the traces for 400 W plasma power, where the propane was also shut off.

In Figure 13 there are two traces for a given power level with the exception of 100 W. The purpose was to determine reproducibility of the startup, which indeed it is.

The turn off time is comparable to the turn on time, indicating very fast turn-on of the plasmatron. It is likely that the rate limitations in the transients in this paper are instrumental, as discussed in the methane start-up paper [2]. It is possible to utilize Figure 13 to evaluate the time constant for the turn-off. The e-folding time is about 0.5 s, thus the time for 90% change (2 e-foldings) is about 1 s

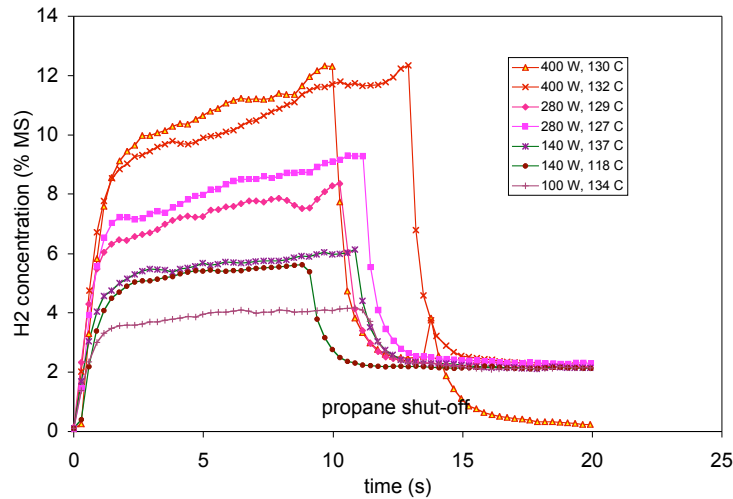


Figure 13. Transient characteristics for premixed propane/swirl air for different values of power, for $O/C \sim 1.3$.

6. Stoichiometric combustion propane/air mixture for swirl gas

The previous section discussed the case of premixed/swirl air. Because the overall O/C is below stoichiometric combustion, the O/C ratio of the propane/swirl air is very low, much lower than 1. The results of making the propane/swirl air prone to igniting because of operation at stoichiometric combustion, with the rest of the propane premixed with the wall air, is discussed in this section. In order to be able to premix the propane with both the swirl air and the wall air, two independent propane controls are required. Two similar rotameters were used to accomplish this.

Results of the experiments are shown in Figure 14. The maximum hydrogen concentration is about 12% at an optimum $O/C \sim 1.5$. The efficiency at the optimal conditions is on the order of 50%. Carbon balance is particularly bad at the lowest O/C ratios in Figure 14, but moderately good ($\sim 10\%$) elsewhere.

The transient hydrogen concentration and temperature are shown in Figure 15 for several power levels. Two traces are shown for each power level. The transient looks similar to those other with low values of O/C , with the prompt hydrogen concentration being a function of plasma power. For the lowest value of power, 50 W, startup is not reproducible, and one of the traces shows a failed startup, while the other shows unstable operation with a frequency of about 0.3 Hz. In this case, as the plasma power is shut off, the hydrogen concentration drops to about 6%. Even for 100 W, the hydrogen concentration is not affected by shutting off the plasma.

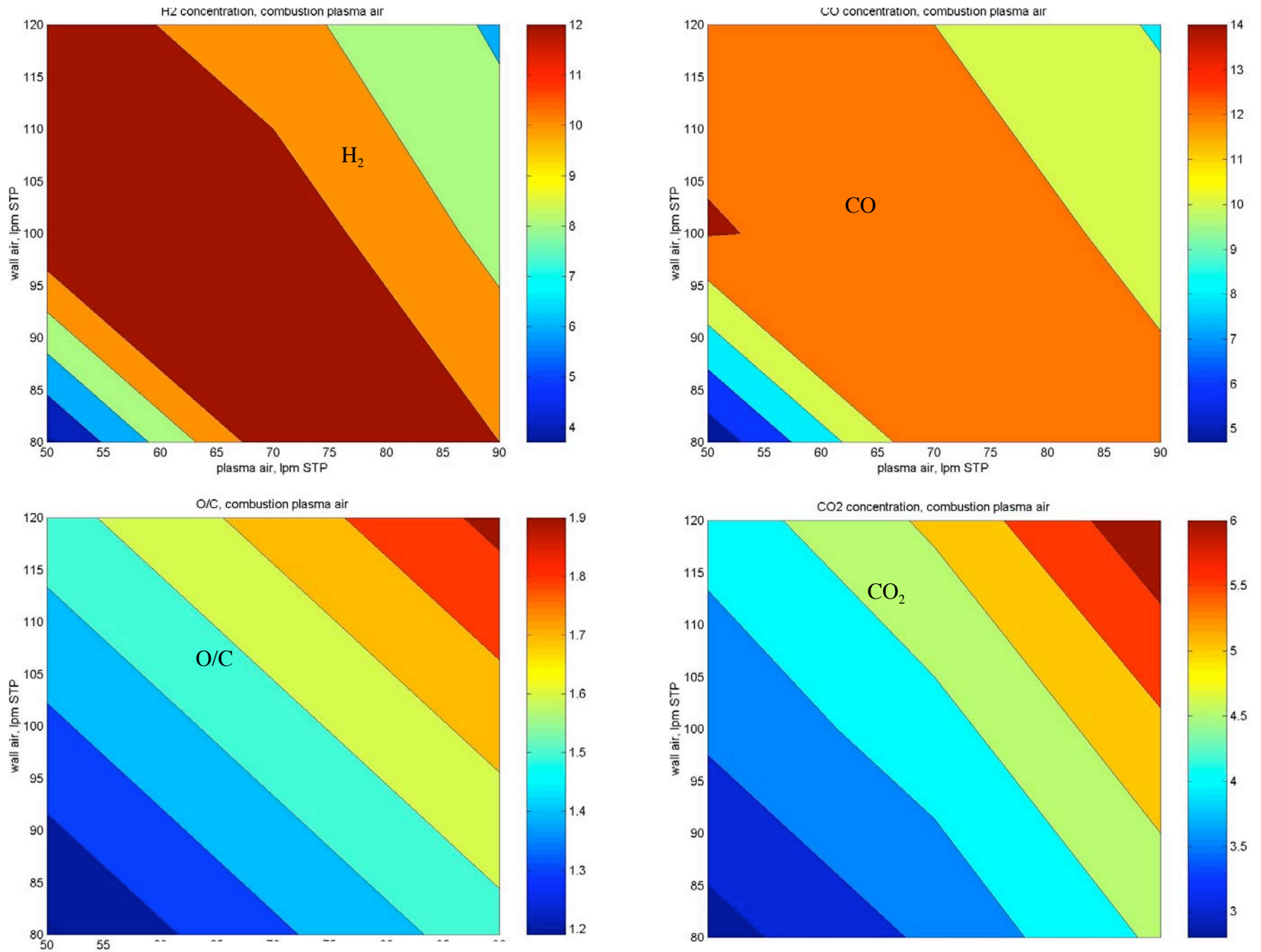


Figure 14. Contours of constant (a) H₂ (b) CO (c) O/C ratio (d) CO₂ as a function of wall and swirl air flow rates, for propane/swirl air at stoichiometric combustion composition, with the rest of propane premixed will wall air.

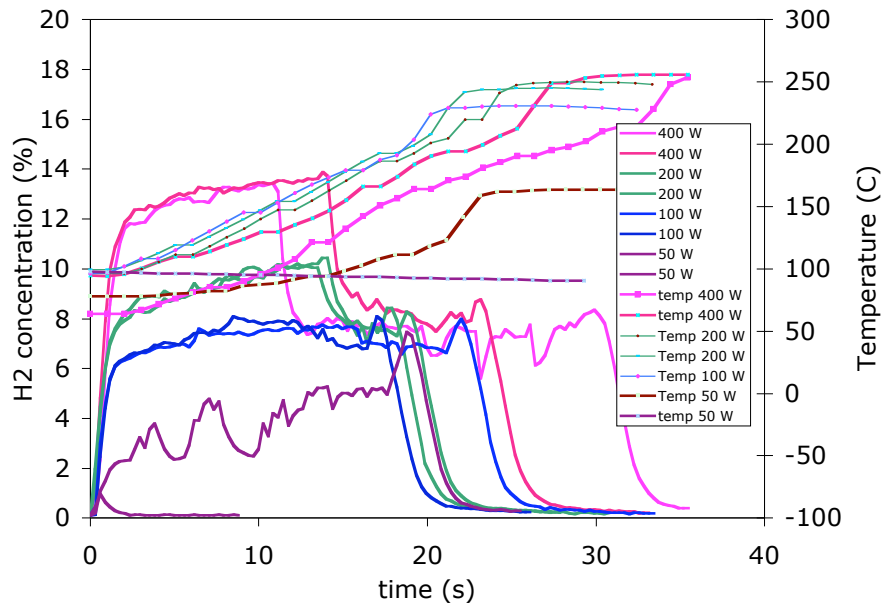


Figure 15. Transients for the case of stoichiometric combustion composition of the swirl gas, with the rest of propane pre-mixed with the wall air, for several values of plasma power.

7. Fully premixed propane and air.

In the last case investigated, the propane and the air are fully premixed. In this case, the plasmatron air/fuel mixture is homogeneous, as all the inputs have the same composition.

Results are presented in Figure 16. Hydrogen concentration is about 12%, at an O/C \sim 1.7. The higher value of the O/C ratio results in increased CO₂ concentration (4.5%), higher temperatures (850-880 C) and lower efficiency (55-60%).

Under these conditions the hydrogen concentration was a strong function of power, with the reaction stopping at slightly decreased values of power (300W).

Figure 17 shows the transients for two levels of power. At 150 W, the reaction sputters but at 350 W the prompt hydrogen concentration is about 8%, with a relatively high rate of rise of the temperature.

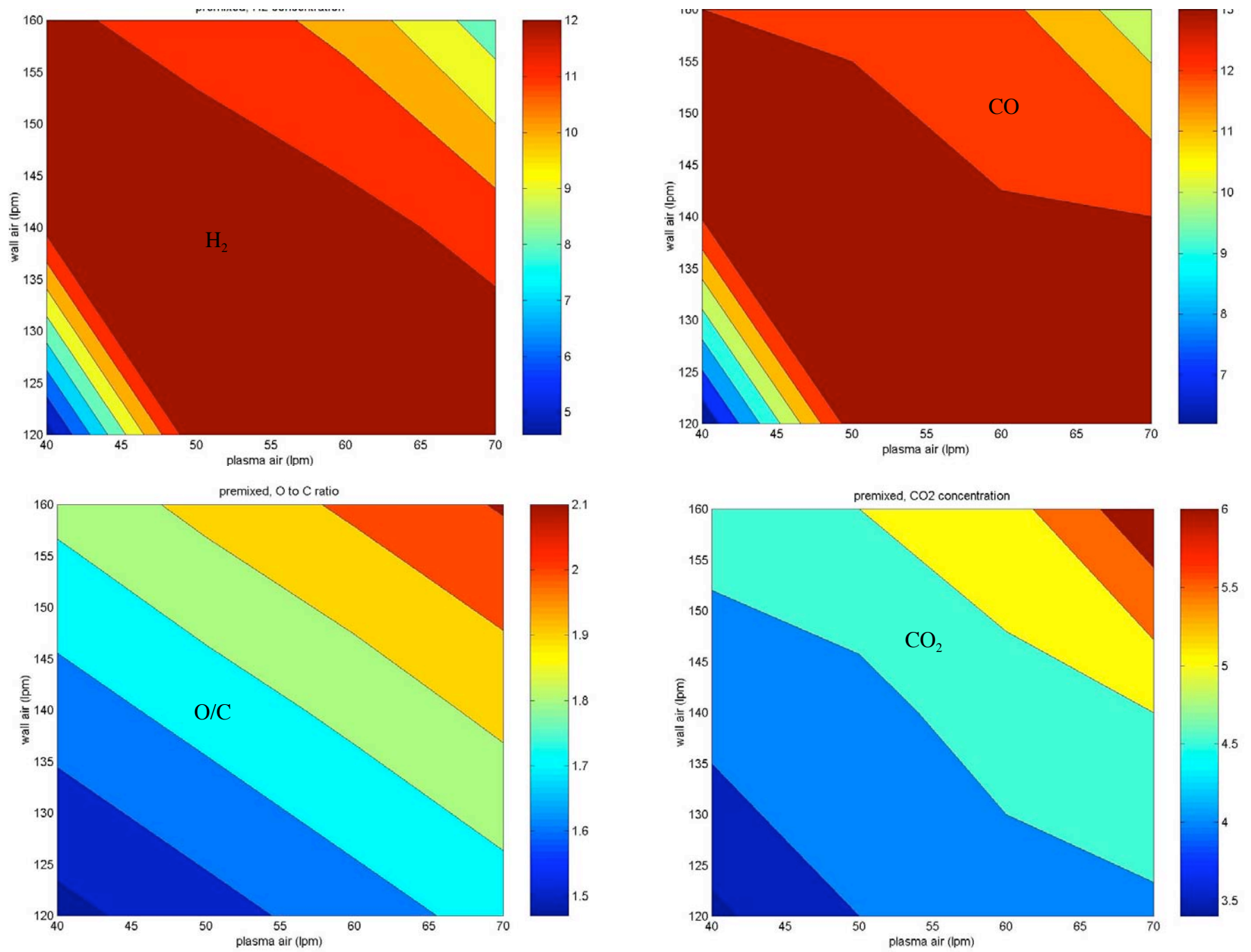


Figure 16. Contours of constant (a) H₂ (b) CO (c) O/C ratio (d) CO₂ as a function of wall and swirl air flow rates, for propane premixed with air, thus resulting in a homogeneous air/propane mixture throughout the device

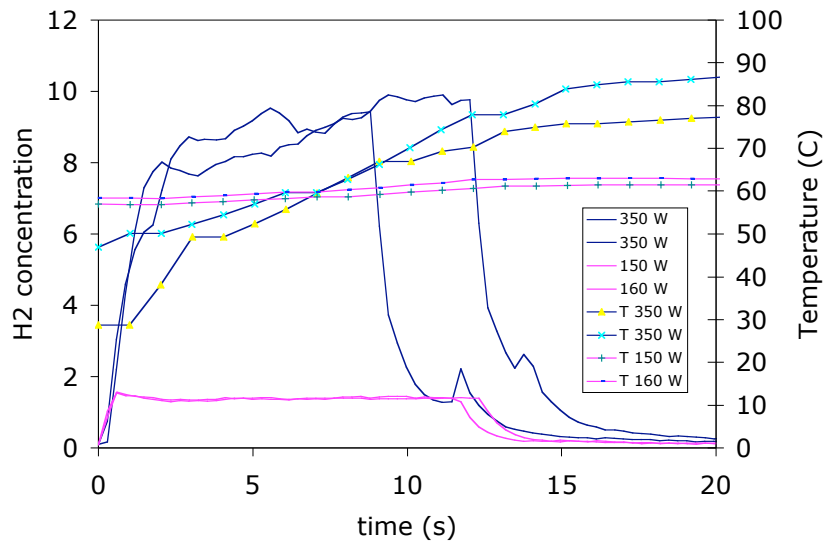


Figure 17. Transients for the case of propane/air premixed, resulting in a homogeneous air/fuel mixture.

8. Discussion

This paper has discussed 5 different means of introducing propane into a plasmatron fuel converter. Both the steady state characteristics, as well as some startup properties have been discussed.

Table 2 Summary of tests presented in this paper

	O/C ratio	Flow dist	Efficiency	H2 conc	Temp	Dependence on power	Cold start up
Premixed	1.7	140/60	60%	12%	850	> 350 W	8% in 1.5 s
Premixed plasma	1.3	75/70	75%	14.50%	720	> 100W	10% in 2 s Continues ~2% when plasma off
Axial	1.8	160/65	65%	12%	790		8% in 1.5 s
Premixed wall	1.6	110/70	55%	13%	820	Drops 20% when hot, plasma off	8% in 1.5 s Power > 150 W
Combustion plasma air	1.45	95/70	60%	12%	800	Drops by 10-20% when plasma off	10% in 1.5 s Power > 100 W

Table 2 shows a summary of the work presented in this paper. Conditions of optimal reforming (highest hydrogen concentration) for each case are shown. It is clear that the results are best when the propane is introduced through the swirl port (premixed plasma)

with the highest values of hydrogen concentration with lowest values of CO₂ and temperature (a sign of combustion). Introducing a stoichiometric for combustion mixture in the swirl gas is not better than when all the propane is introduced through the swirl port, but it is better than the other configurations. Some of the results from the fluid dynamic simulation in section 2 suggest that premixing the propane with the swirl air results in the highest fraction of propane in stoichiometric combustion conditions in the plasma region.

It is interesting to note that the worst configuration is that with the fuel injected through the axial port. Unfortunately, this is the configuration that is needed for injection of liquid fuels.

The plasmatron configuration that was chosen for the experiments was designed for conversion of diesel fuel. There is a long path between the axial injection point and the electrode gap, where the plasma is. It is in this region that the axially injected propane premixes with the wall air. For liquid fuels, which require air assist for atomization, the velocity of the fuel droplets is very high, and it is possible to maintain stratification even when axially injecting the fuel.

9. Conclusion

In general, the following conclusions can be derived from this work:

- O/C controls the performance of the device, more than where the air is introduced (swirl air, wall air),
- The device is sensitive to the place of introduction of the propane, with best results premixing the propane with the swirl air and the worst results with the propane injected through the axial nozzle.
- Reforming is sensitive to plasma power at lower values of O/C. At higher values of O/C, the plasma is mainly needed for startup.
- Startup is sensitive to power at lower values of O/C.

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