

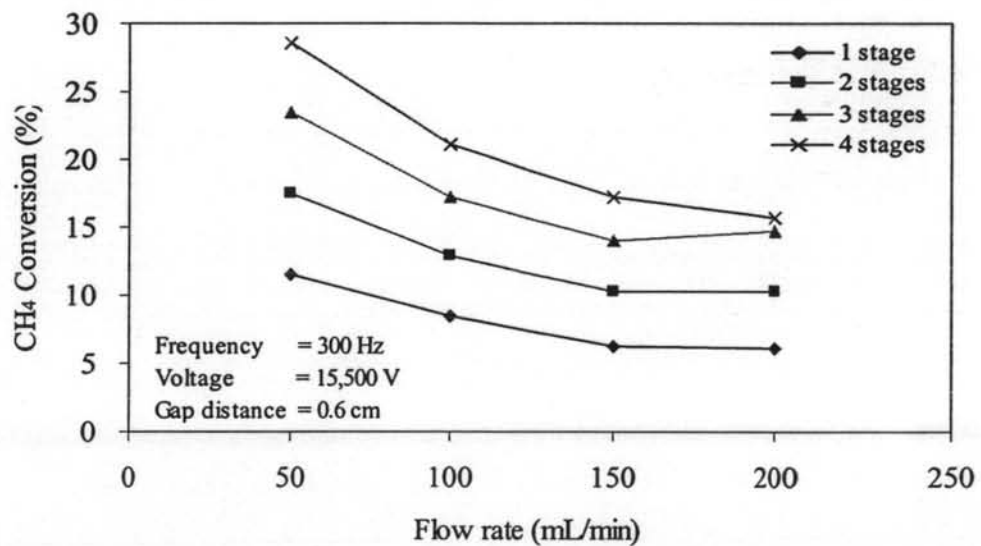
## CHAPTER IV

### RESULTS AND DISCUSSION

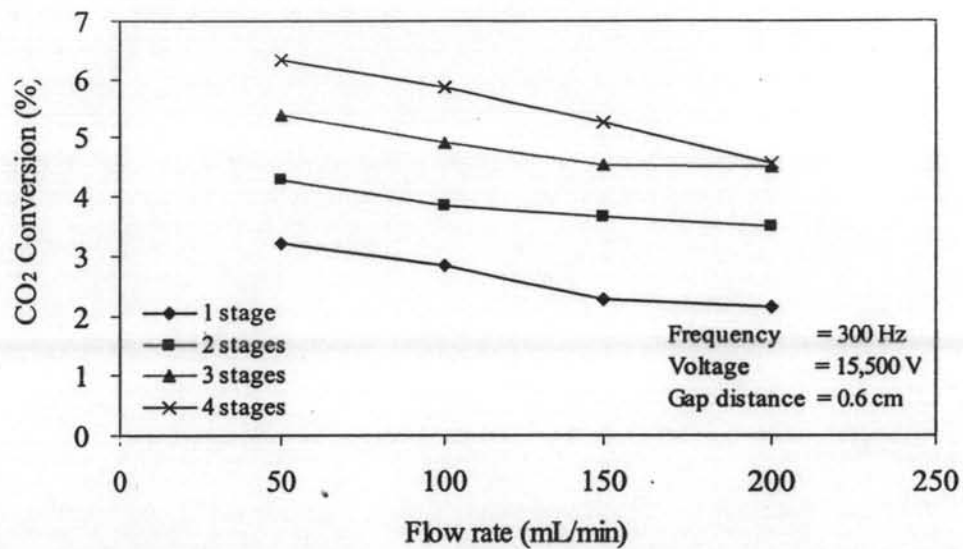
#### 4.1 Effect of Feed Flow Rate

##### 4.1.1 Effect on Methane and Carbon Dioxide Conversion

The effects of feed flow rate on  $\text{CH}_4$  and  $\text{CO}_2$  conversions are illustrated in Figures 4.1 and 4.2, respectively. Feed flow rate is one of the most important operating parameters affecting the performance of the plasma system because it directly relates to residence time. Both  $\text{CH}_4$  and  $\text{CO}_2$  conversions decreased with increasing feed flow rate from 50-200 mL/min. This result leads to a conclusion that either the feed flow rate increases or the stage number decreases resulting in decreasing residence time. Therefore the contact time of methane and carbon dioxide molecules with electrons decreases leading to lower conversion of both  $\text{CH}_4$  and  $\text{CO}_2$ .



**Figure 4.1** Effect of feed flow rate on  $\text{CH}_4$  conversion at different stage numbers of the plasma system.



**Figure 4.2** Effect of feed flow rate on CO<sub>2</sub> conversion at different stage numbers of the plasma system.

#### 4.1.2 Effect on Product Selectivities

Figures 4.3 to 4.7 show the effect of feed flow rate on the selectivities of H<sub>2</sub>, CO, C<sub>2</sub>H<sub>2</sub>, C<sub>2</sub>H<sub>4</sub> and C<sub>2</sub>H<sub>6</sub> respectively. As the feed flow rate increased, the C<sub>2</sub>H<sub>6</sub> selectivity slightly increased, in the contrast the selectivities of H<sub>2</sub>, CO, C<sub>2</sub>H<sub>2</sub> and C<sub>2</sub>H<sub>4</sub> decreased. The results can be explained that the feed flow rate was increased leading to decreasing residence time. As a result, more ethane molecules will not dehydrogenation then ethylene and acetylene molecules are decreased.

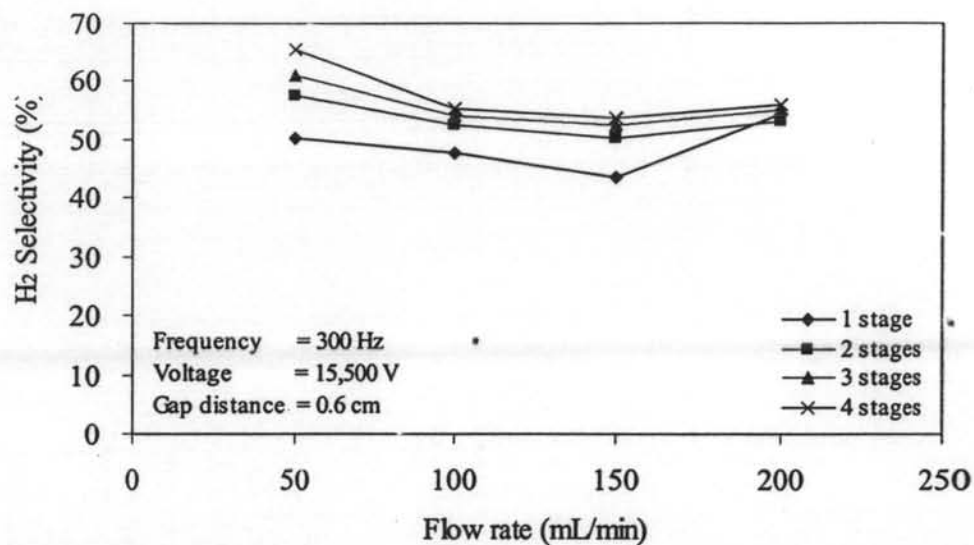


Figure 4.3 Effect of feed flow rate on H<sub>2</sub> selectivity at different stage numbers of the plasma system.

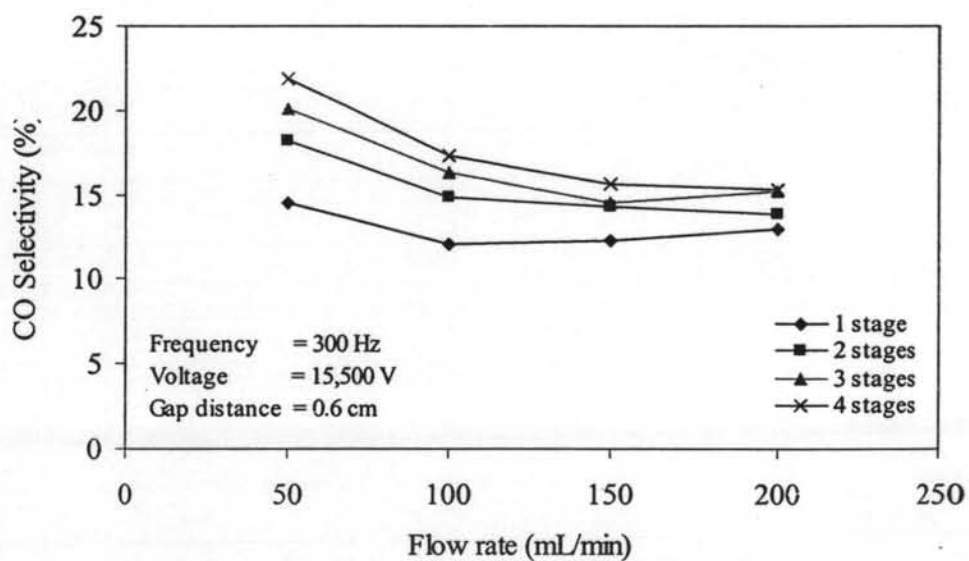
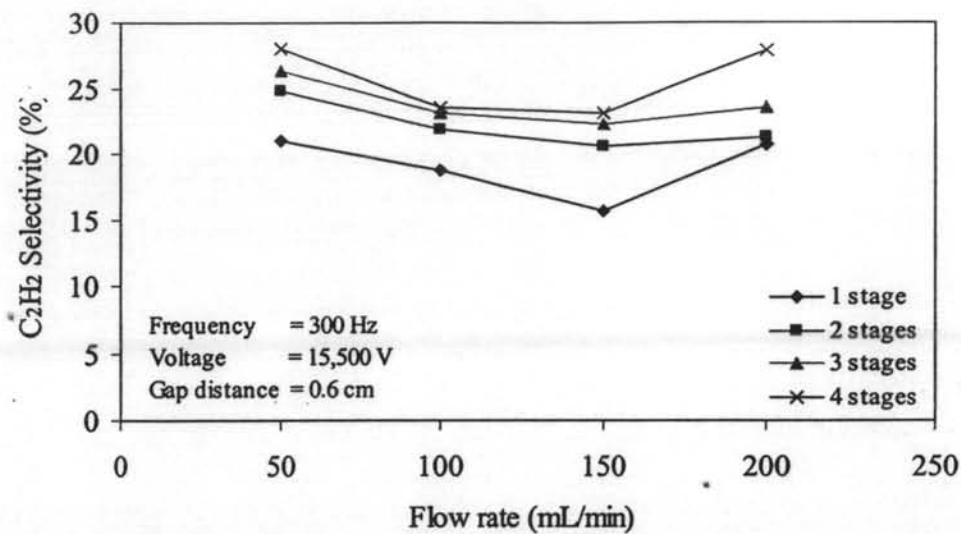
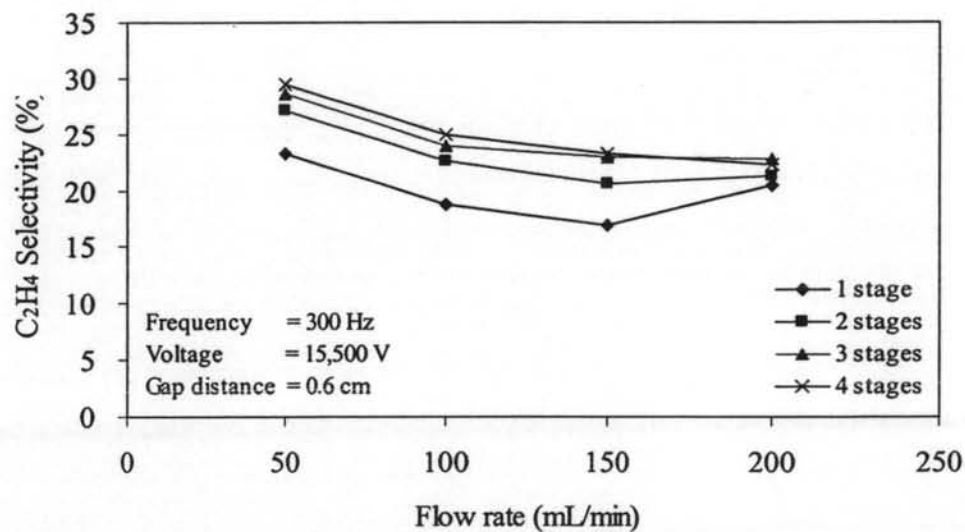


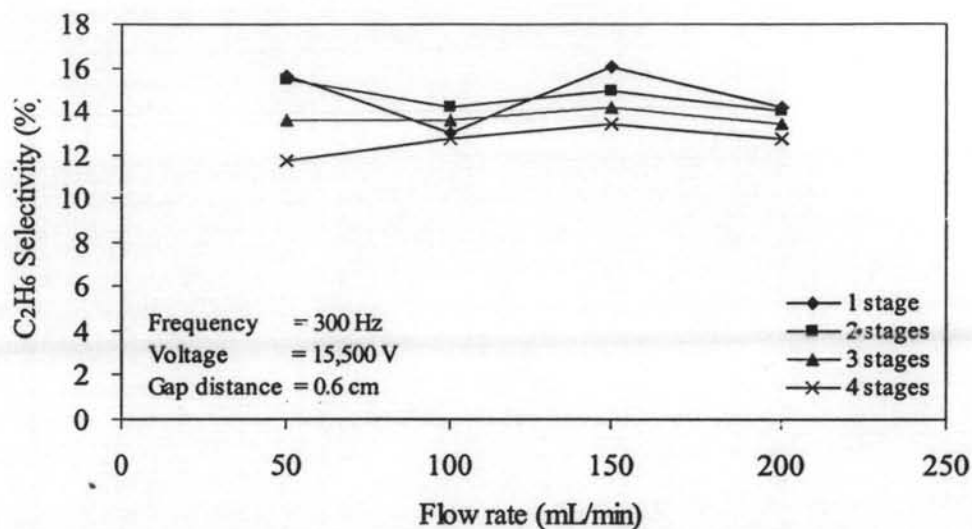
Figure 4.4 Effect of feed flow rate on CO selectivity at different stage numbers of the plasma system.



**Figure 4.5** Effect of feed flow rate on  $C_2H_2$  selectivity at different stage numbers of the plasma system.



**Figure 4.6** Effect of feed flow rate on  $C_2H_4$  selectivity at different stage numbers of the plasma system.

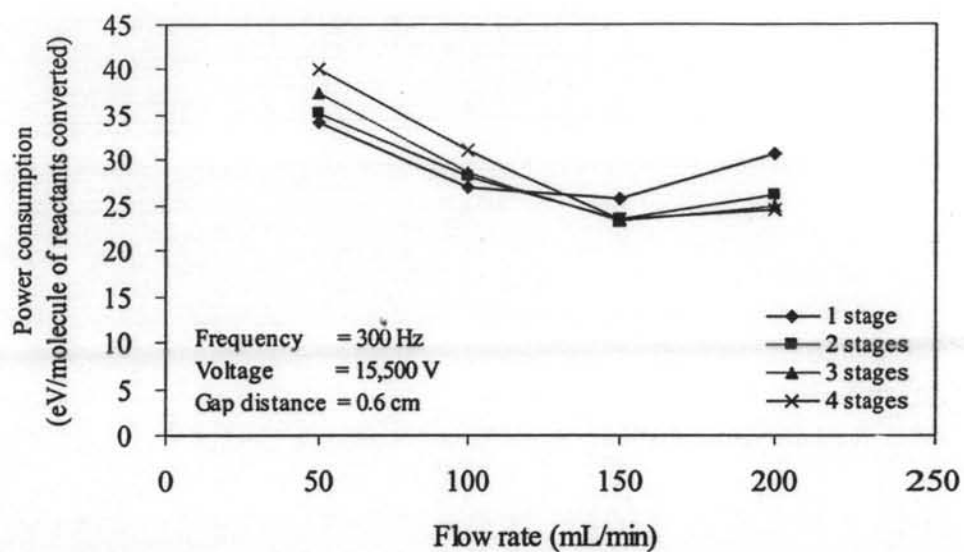


**Figure 4.7** Effect of feed flow rate on  $C_2H_6$  selectivity at different stage numbers of the plasma system.

#### 4.1.3 Effect on Power Consumption

As can be seen from Figure 4.8, the minimum power consumption is approximately achieved at the feed flow rate about 150-200 mL/min. At a flow rate lower than 150 mL/min, power consumption increased with decreasing feed flow rate because of lower probabilities of reacting with lower amounts of reactants available and some of the power may be consumed by secondary reaction. (Supat *et al.*, 2003)

According to the power consumption, a feed flow rate in the range of 150-200 mL/min should be considered for operating this plasma reactor system. Even though the system has the low power consumption at a feed flow rate of 200 mL/min, it had a lower methane conversion as compared to that operated at a feed flow rate 150 mL/min. As a results, a feed flow rate of 150 mL/min was selected to operate the system to determine other parameters.

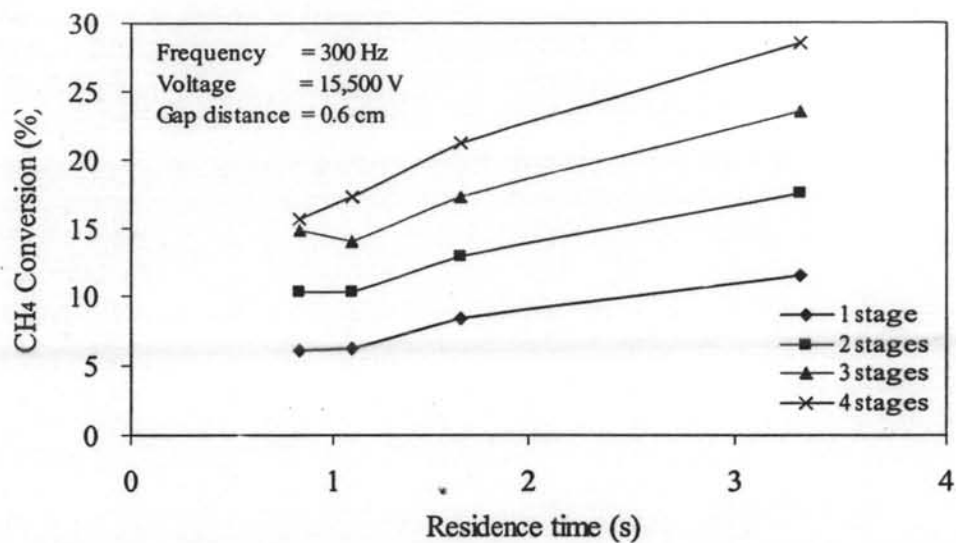


**Figure 4.8** Effect of feed flow rate on power consumption at different stage numbers of the plasma system.

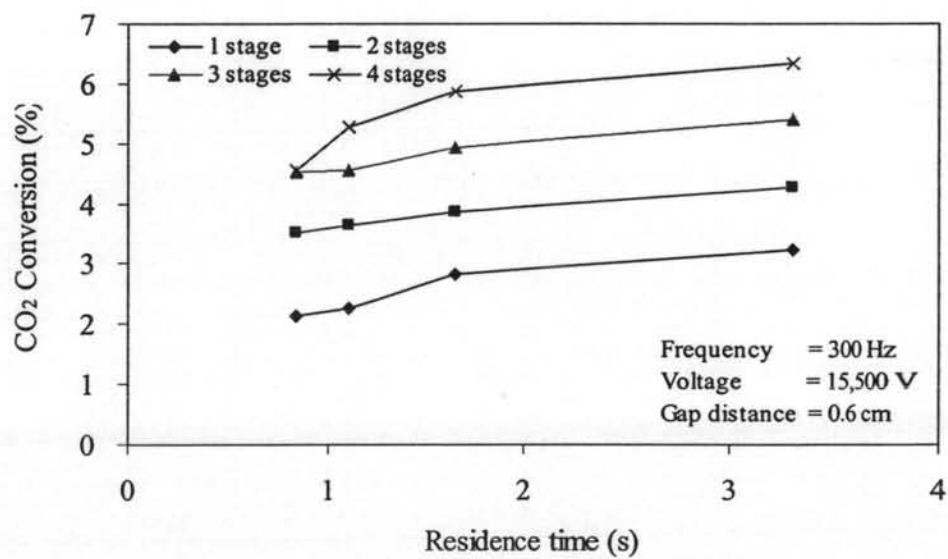
## 4.2 Effect of Residence Time

### 4.2.1 Effect on Methane and Carbon Dioxide Conversion

The effect of residence time on the process performance of the gliding arc system was studied by varying the feed flow rate from 50 to 200 mL/min corresponding to residence time of 0.83 to 3.31 seconds. The reaction volume was determined from the observation of the plasma appearance in each reactor which was approximately 3 mL for each reactor. The results of methane and carbon dioxide conversions are shown in Figures 4.9 and 4.10, respectively. The conversions of  $\text{CH}_4$  and  $\text{CO}_2$  significantly increased with increasing residence time and stage number. When either the residence time or stage number is increased, a longer contact time of reactant gases passing through the reaction zone is obtained resulting in more chance of both  $\text{CH}_4$  and  $\text{CO}_2$  molecules to be contacted with electrons. As a result, both  $\text{CH}_4$  and  $\text{CO}_2$  conversions increase with increasing residence time.



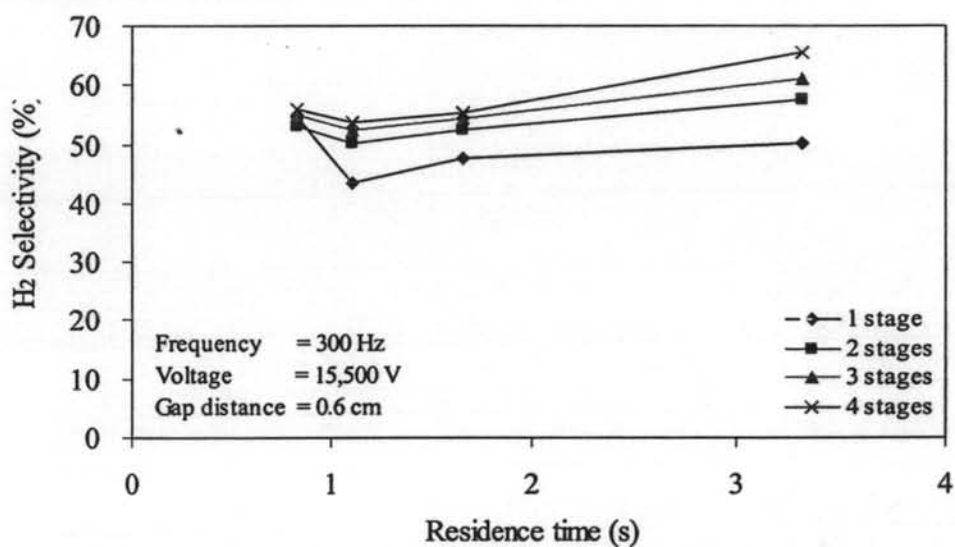
**Figure 4.9** Effect of residence time on CH<sub>4</sub> conversion at different stage numbers of the plasma system.



**Figure 4.10** Effect of residence time on CO<sub>2</sub> conversion at different stage numbers of the plasma system.

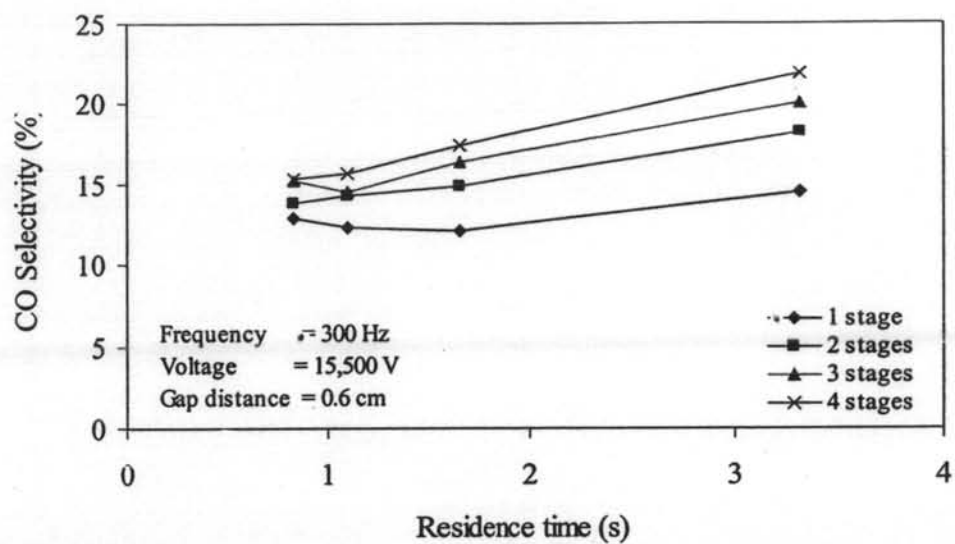
#### 4.2.2 Effect on Product Selectivities

Figures 4.11 to 4.15 show the effect of residence time on the product selectivities. The selectivities of  $H_2$ ,  $CO$ ,  $C_2H_2$  and  $C_2H_4$  slightly increased while the selectivities of  $C_2H_6$  slightly decreased with increasing residence time. The explanation for the effect of residence time is the same as for the effect of feed flow rate.

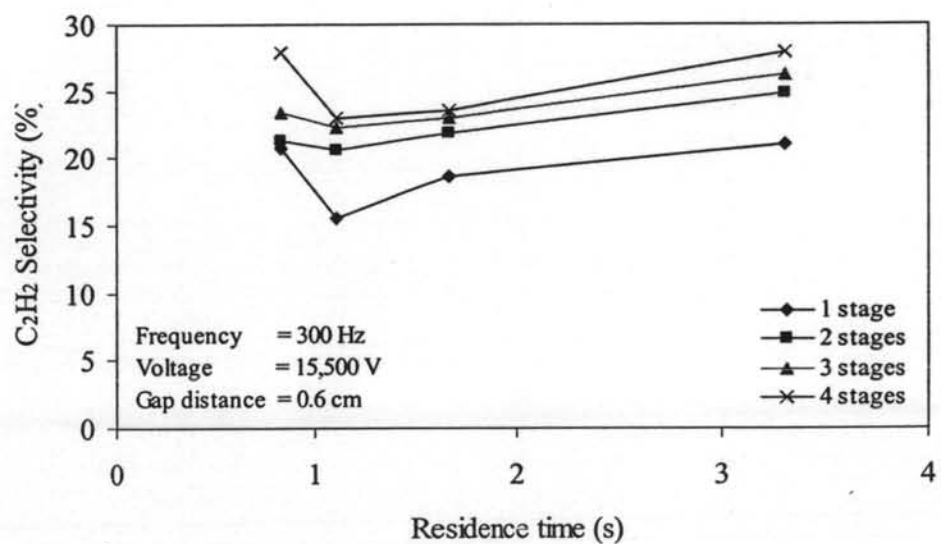


**Figure 4.11** Effect of residence time on  $H_2$  selectivity at different stage numbers of the plasma system.





**Figure 4.12** Effect of residence time on CO selectivity at different stage numbers of the plasma system.



**Figure 4.13** Effect of residence time on C<sub>2</sub>H<sub>2</sub> selectivity at different stage numbers of the plasma system.

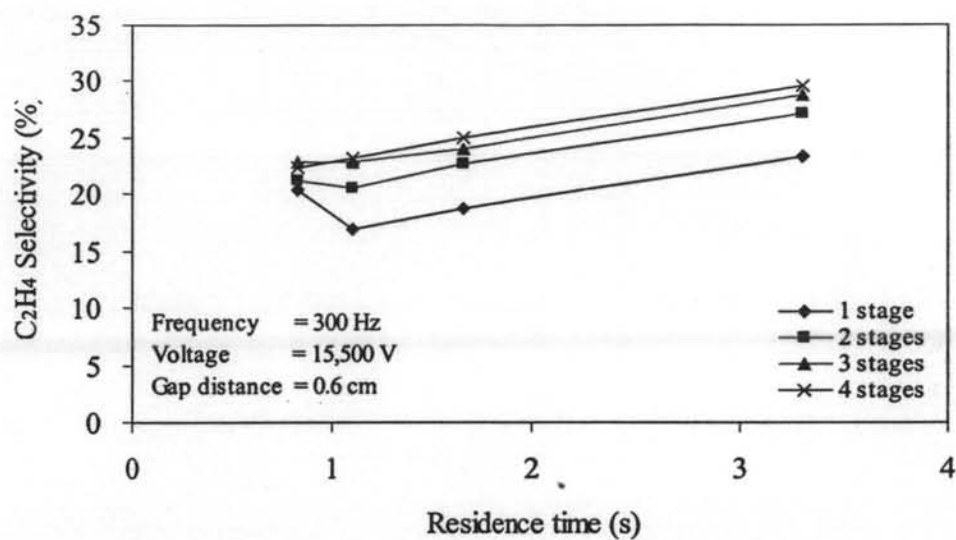


Figure 4.14 Effect of residence time on  $C_2H_4$  selectivity at different stage numbers of the plasma system.

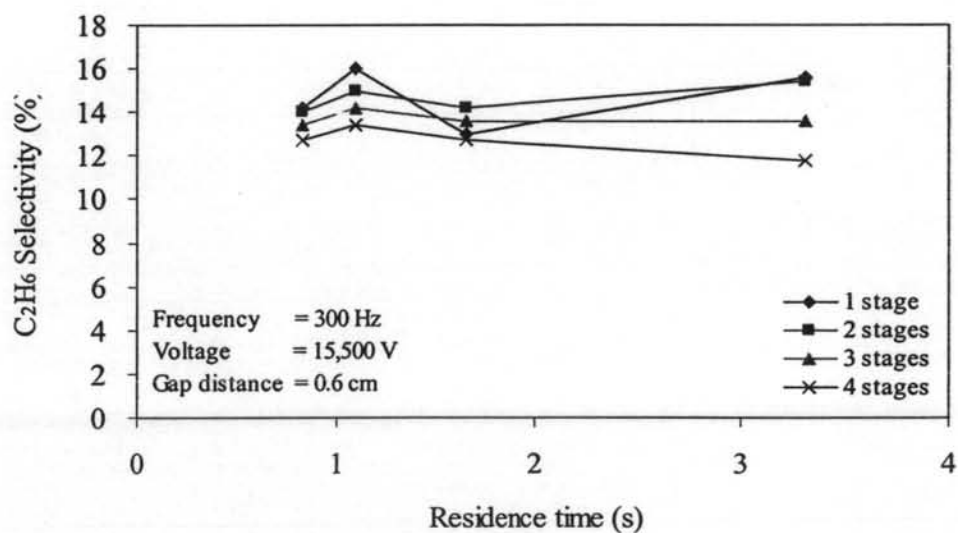


Figure 4.15 Effect of residence time on  $C_2H_6$  selectivity at different stage numbers of the plasma system.

### 4.3 Effect of Frequency

#### 4.3.1 Effect on Methane and Carbon Dioxide Conversion

Figure 4.16 and 4.17 show the effects of frequency on  $\text{CH}_4$  and  $\text{CO}_2$  conversions, respectively, by varying frequency in the range of 300-700 Hz. The system could not be operated at a frequency below 300 Hz because the field is not high enough. The conversions of  $\text{CH}_4$  and  $\text{CO}_2$  decreased with increasing frequency. The main effect of frequency on the conversions results from the space charge (electrons and ions) characteristics of the discharge. At a frequency of 300 Hz with the highest conversions of both  $\text{CH}_4$  and  $\text{CO}_2$ , highest current is at this frequency as show in figure 4.18. At the highest current, there was the largest numbers of electrons generated in the system to initiate the reactions resulting in both maximum  $\text{CH}_4$  and  $\text{CO}_2$  conversions. For any given frequency, both  $\text{CH}_4$  and  $\text{CO}_2$  increased with increasing stage numbers since the residence time increases.

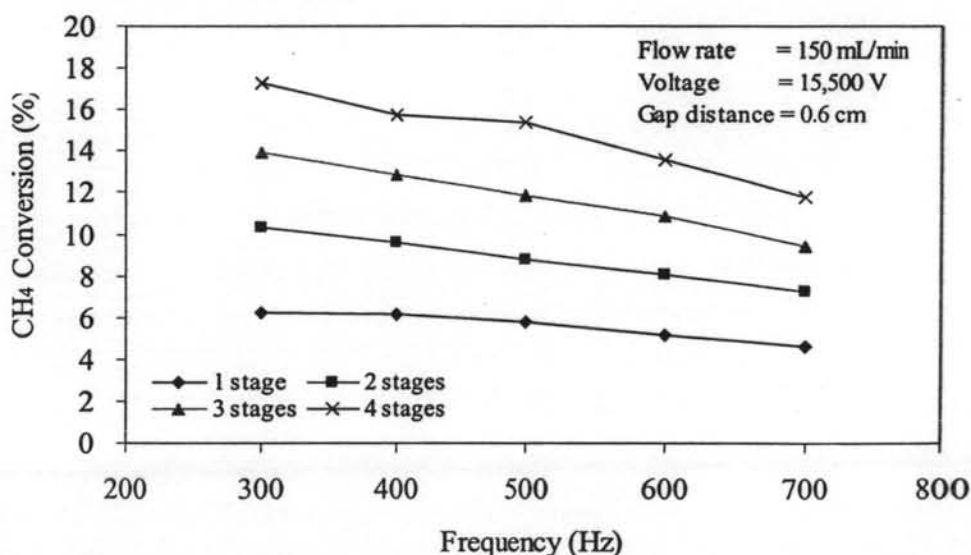


Figure 4.16 Effect of frequency on  $\text{CH}_4$  conversion at different stage numbers of the plasma system.

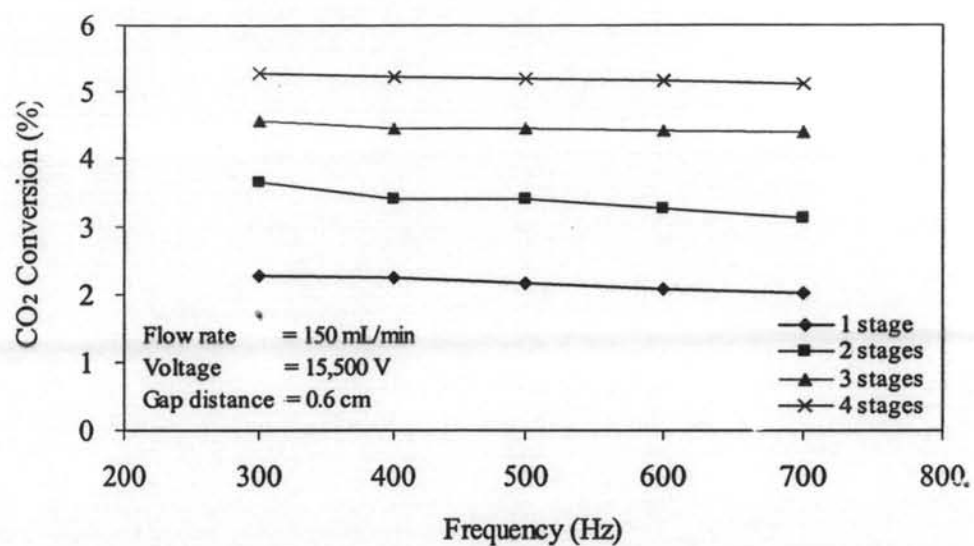


Figure 4.17 Effect of frequency on CO<sub>2</sub> conversion at different stage numbers of the plasma system.

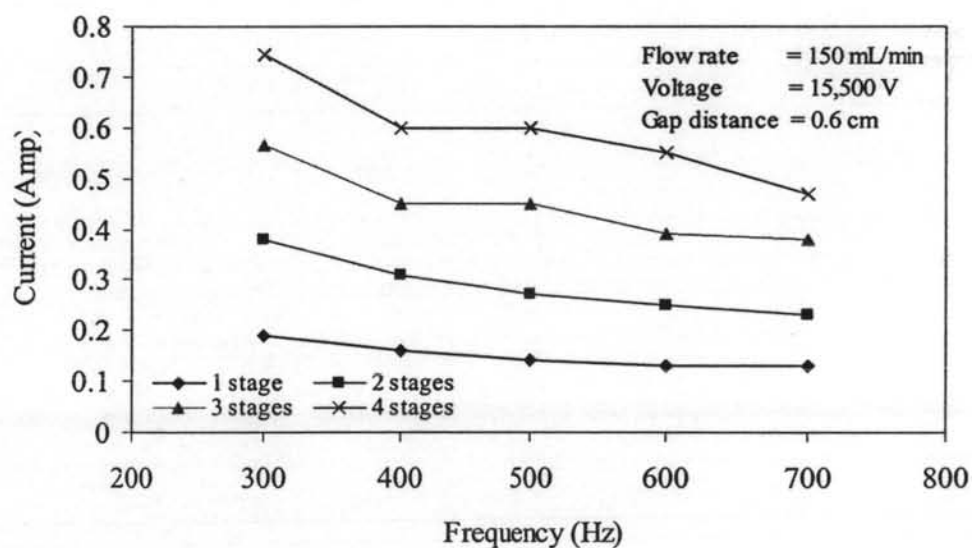


Figure 4.18 Effect of frequency on current at different stage numbers of the plasma system.

#### 4.3.2 Effect on Product Selectivities

The effects of frequency on product selectivities are shown in Figure 4.19 to 4.23. The selectivities of CO and C<sub>2</sub>H<sub>6</sub> increased with increasing frequency but the selectivities of C<sub>2</sub>H<sub>2</sub> and C<sub>2</sub>H<sub>4</sub> decreased while the H<sub>2</sub> selectivity did not change significantly. As mention before, a higher frequency gives a lower current. As a result, there will be less electrons available to initiate the reactions at a higher frequency. The result reveal that the effect of frequency on the biogas reforming reaction to produce synthesis gas is much higher than the coupling reaction of methane and dehydrogenation reaction.

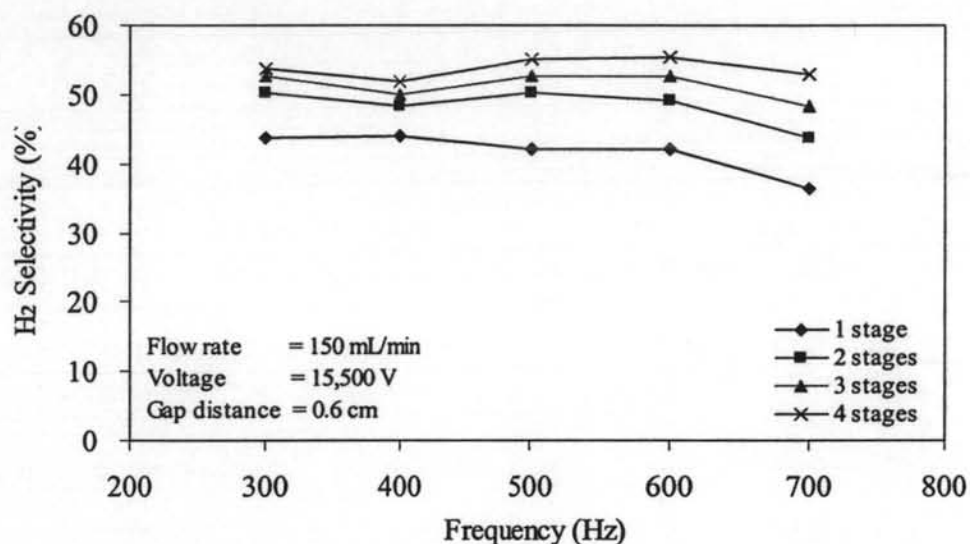


Figure 4.19 Effect of frequency on H<sub>2</sub> selectivity at different stage numbers of the plasma system.

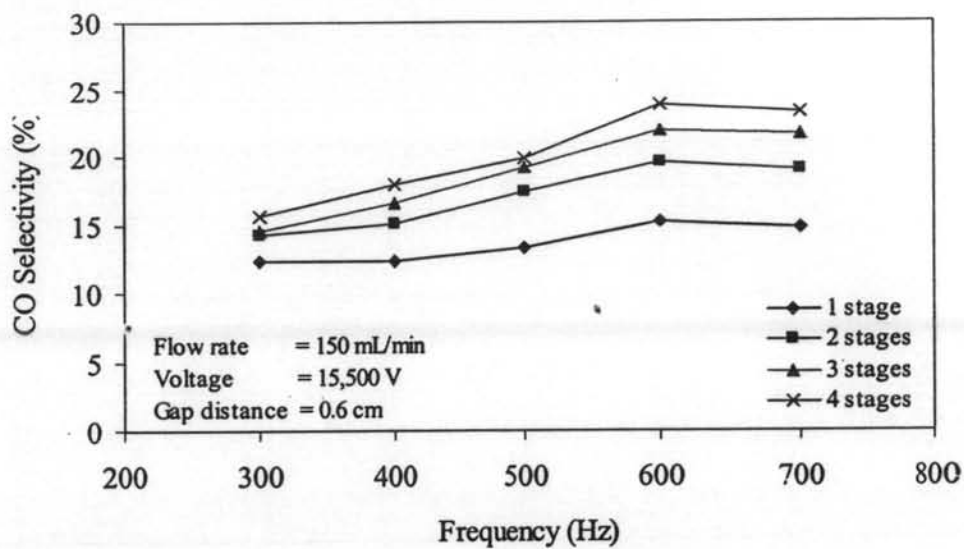


Figure 4.20 Effect of frequency on CO selectivity at different stage numbers of the plasma system.

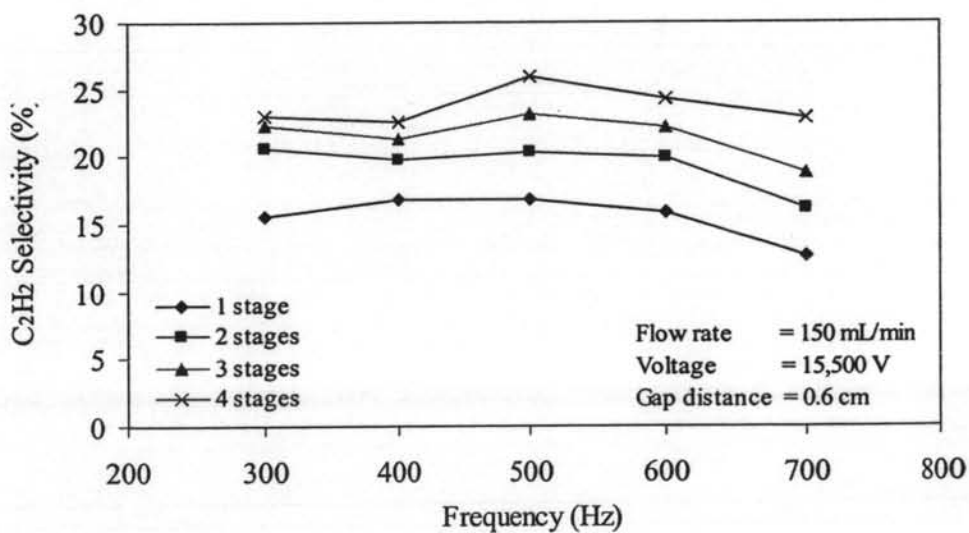


Figure 4.21 Effect of frequency on C<sub>2</sub>H<sub>2</sub> selectivity at different stage numbers of the plasma system.

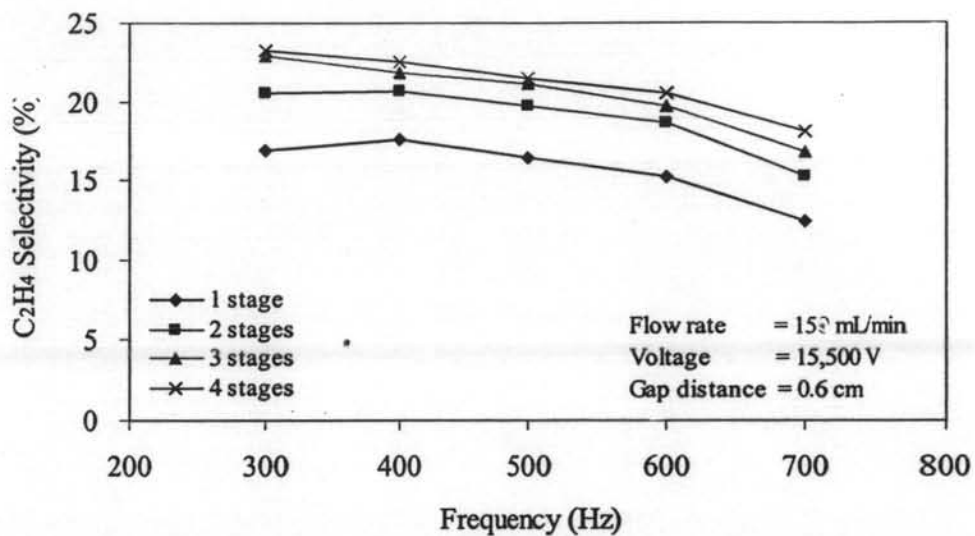


Figure 4.22 Effect of frequency on  $C_2H_4$  selectivity at different stage numbers of the plasma system.

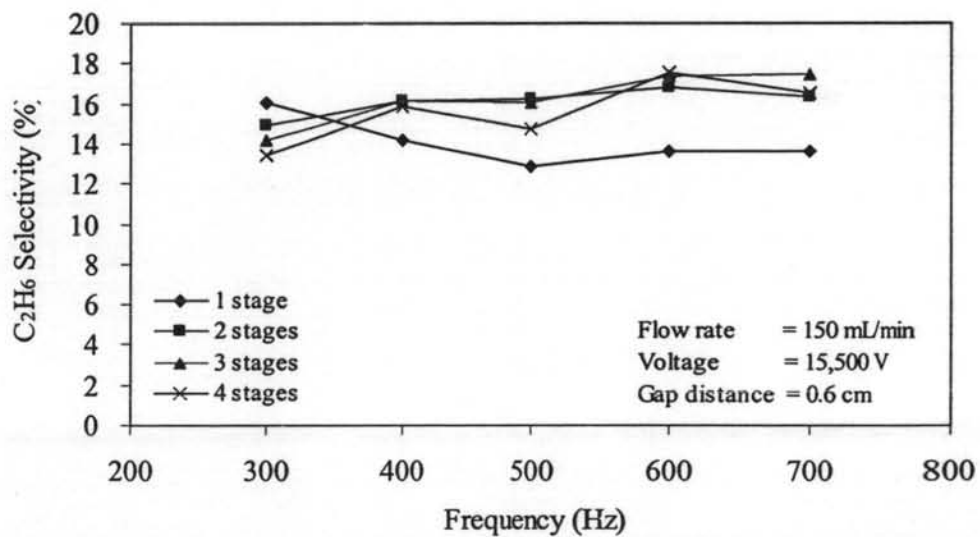


Figure 4.23 Effect of frequency on  $C_2H_6$  selectivity at different stage numbers of the plasma system.

### 4.3.3 Effect on Power Consumption

The effect of frequency on power consumption is illustrated in Figure 4.24. The minimum power consumption was found at a frequency of 500 Hz. However, 300 Hz is selected for next experiments because the highest  $\text{CH}_4$  and  $\text{CO}_2$  conversions can be achieved and the power consumption at 300 Hz is not much different from the power consumption at 500 Hz.

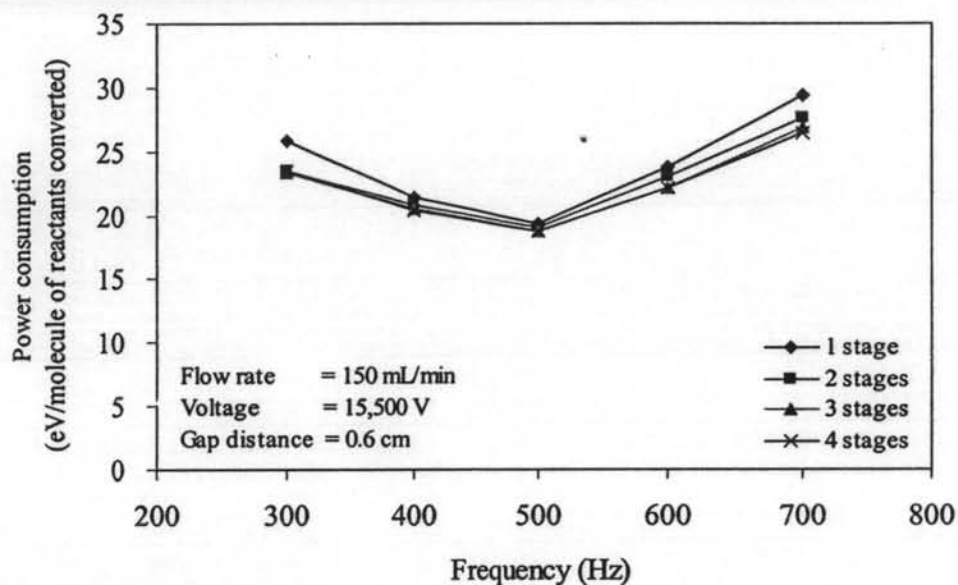


Figure 4.24 Effect of frequency on power consumption at different stage numbers of the plasma system.

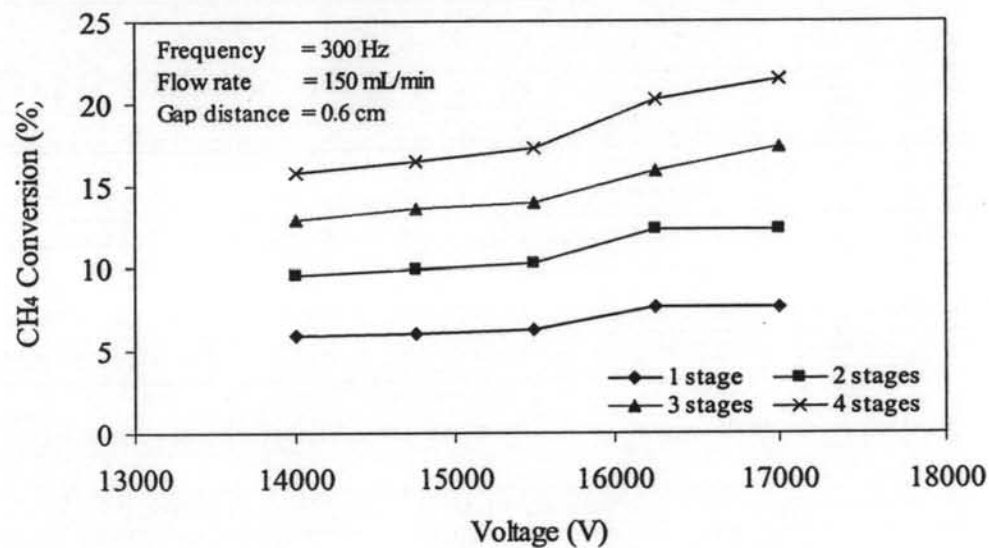
## 4.4 Effect of Applied Voltage

### 4.4.1 Effect on Methane and Carbon Dioxide Conversions

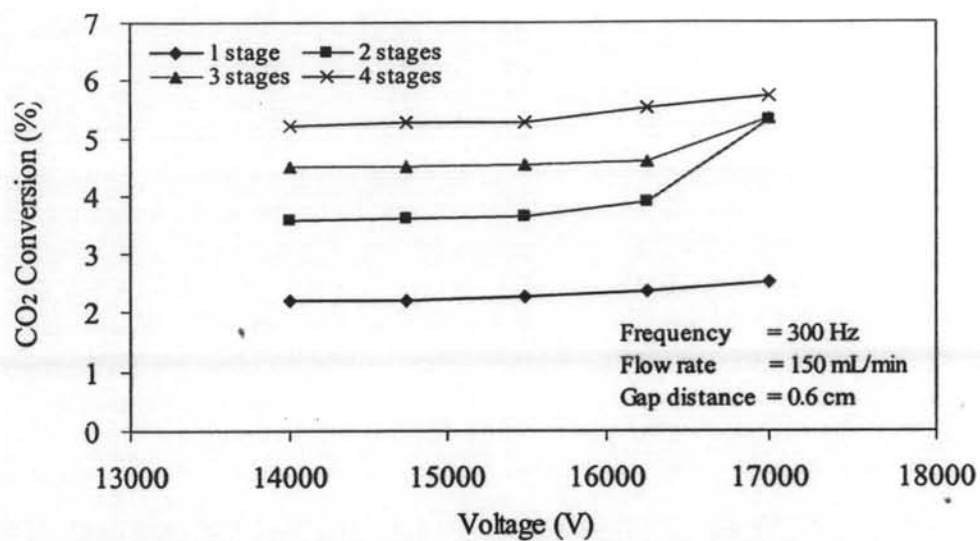
The effects of applied voltage on  $\text{CH}_4$  and  $\text{CO}_2$  conversions are shown in Figures 4.25 and 4.26, respectively. The applied voltage was varied from 14,000 to 17,000 V of the high side voltage since the break-down voltage of the study system is 14,000 V. For an applied voltage greater than 17,000 V, the system could not be operated because of the carbon deposition on the electrodes surfaces. In the



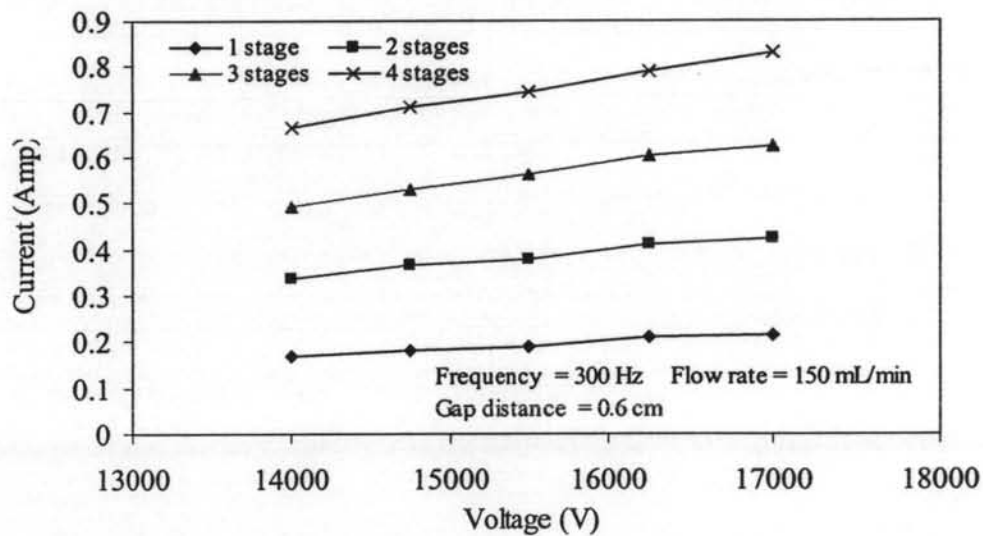
contrast with increasing frequency, the  $\text{CH}_4$  and  $\text{CO}_2$  conversions increased with increasing applied voltage. The explanation is that a higher voltage results in higher current as shown in Figure 4.27. A higher current will give more available electrons to initiate the reactions because of the more opportunity of collision between  $\text{CH}_4$  and  $\text{CO}_2$  with electrons. Morinaga and Suzuki (1962) also found that, with fixed geometry, the quantity of electric transfer between electrodes increased, i.e. the current increased with increasing applied voltage. For any applied voltage, both  $\text{CH}_4$  and  $\text{CO}_2$  conversions increased with increasing the stage number, as a result of increasing residence time. When the residence time increases the collision of  $\text{CH}_4$  and  $\text{CO}_2$  molecules with the sufficient energy electrons increase.



**Figure 4.25** Effect of applied voltage on  $\text{CH}_4$  conversion at different stage numbers of the plasma system.



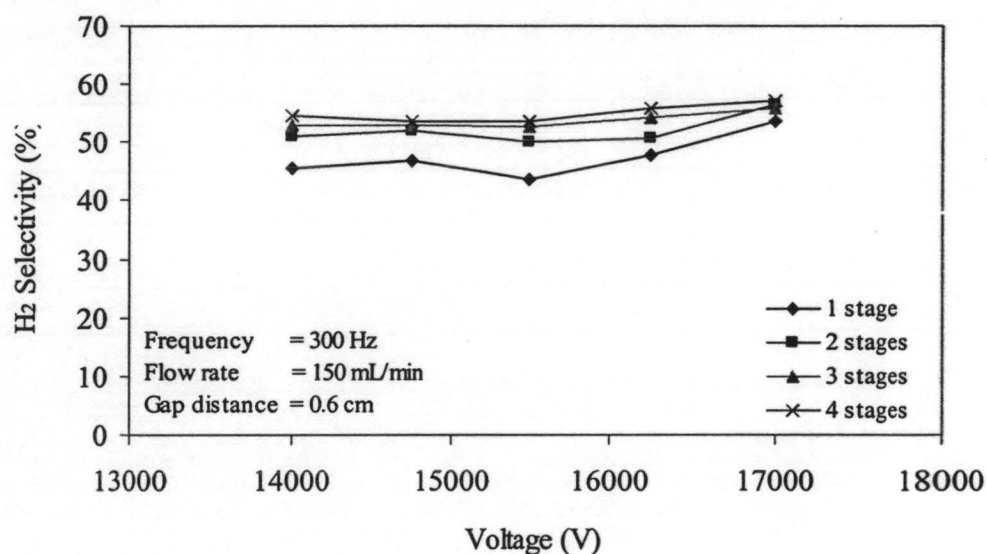
**Figure 4.26** Effect of applied voltage on CO<sub>2</sub> conversion at different stage numbers of the plasma system.



**Figure 4.27** Effect of applied voltage on current at different stage numbers of the plasma system.

#### 4.4.2 Effect on Product Selectivities

The effects of applied voltage on product selectivities are shown in Figures 4.28 to 4.32. The selectivities of CO and C<sub>2</sub>H<sub>6</sub> decreased with increasing applied voltage and stage number, in contrast with the selectivity of C<sub>2</sub>H<sub>2</sub>. The results imply that CO, H<sub>2</sub>, C<sub>2</sub>H<sub>4</sub> and C<sub>2</sub>H<sub>6</sub> are the primary products. When the applied voltage increases leading to higher average electron. Therefore the hydrocarbon primary products (C<sub>2</sub>H<sub>4</sub> and C<sub>2</sub>H<sub>6</sub>) are further reacted known as dehydrogenation to form C<sub>2</sub>H<sub>2</sub> and the CO oxidation is also increased. The oxidation reactions of CO and H<sub>2</sub> produce CO<sub>2</sub>. An increase in the stage number corresponding to a higher residence time enhances both the CO oxidation and dehydrogenation reactions.



**Figure 4.28** Effect of applied voltage on H<sub>2</sub> selectivity at different stage numbers of the plasma system.

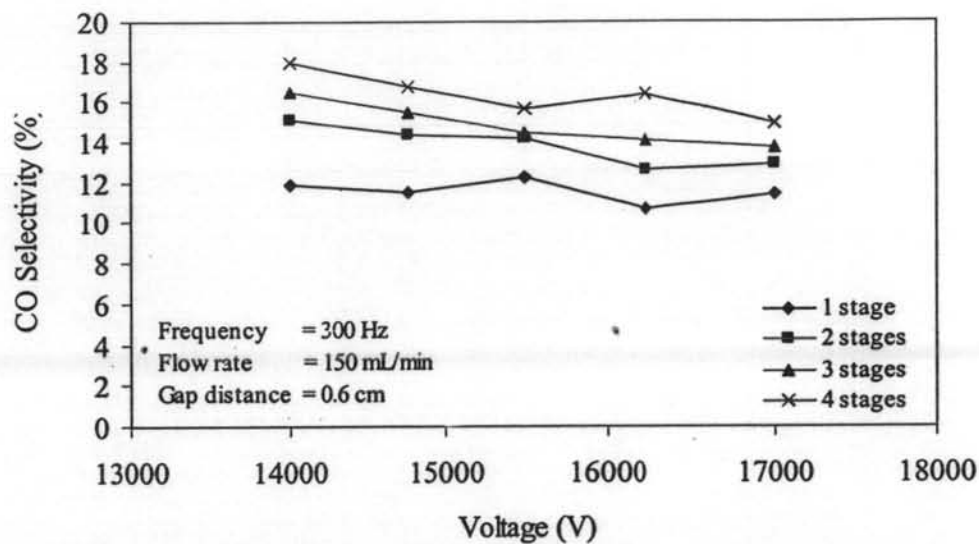


Figure 4.29 Effect of applied voltage on CO selectivity at different stage numbers of the plasma system.

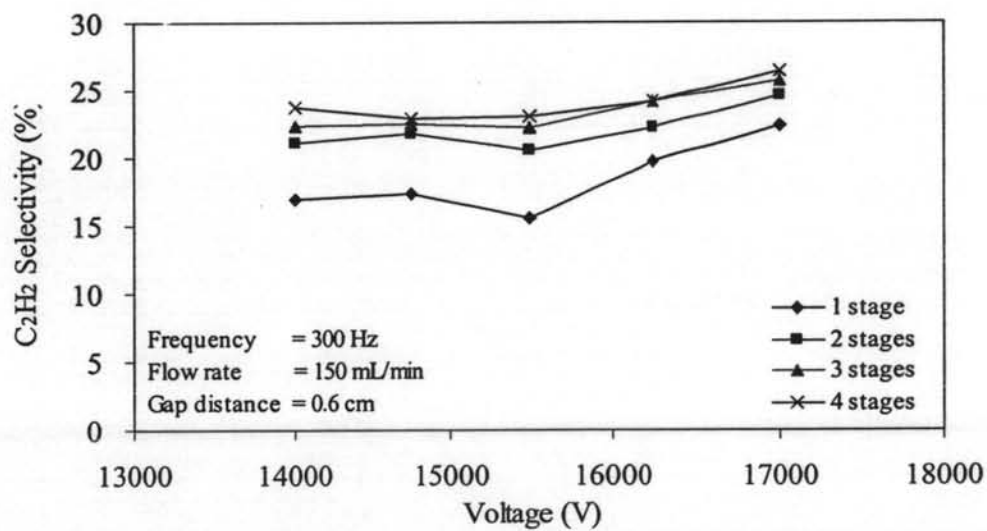


Figure 4.30 Effect of applied voltage on C<sub>2</sub>H<sub>2</sub> selectivity at different stage numbers of the plasma system.

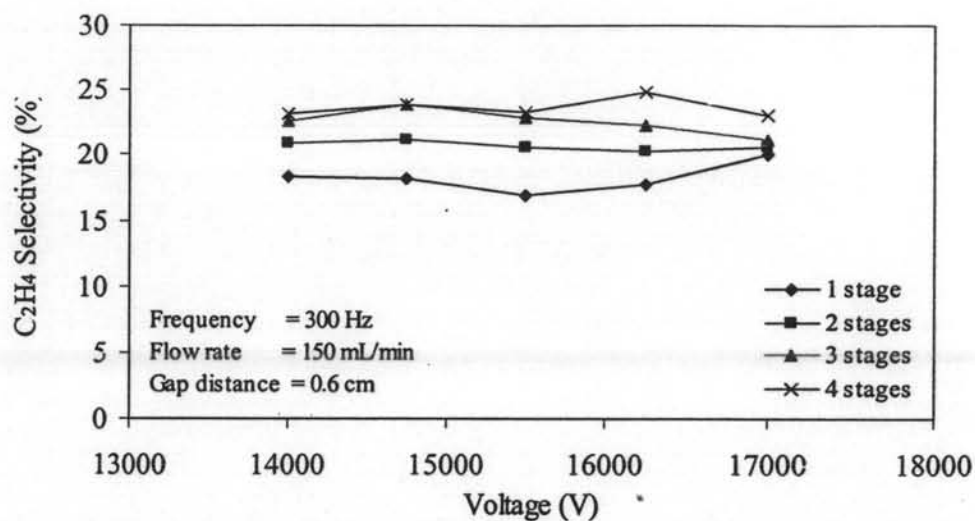


Figure 4.31 Effect of applied voltage on C<sub>2</sub>H<sub>4</sub> selectivity at different stage numbers of the plasma system.

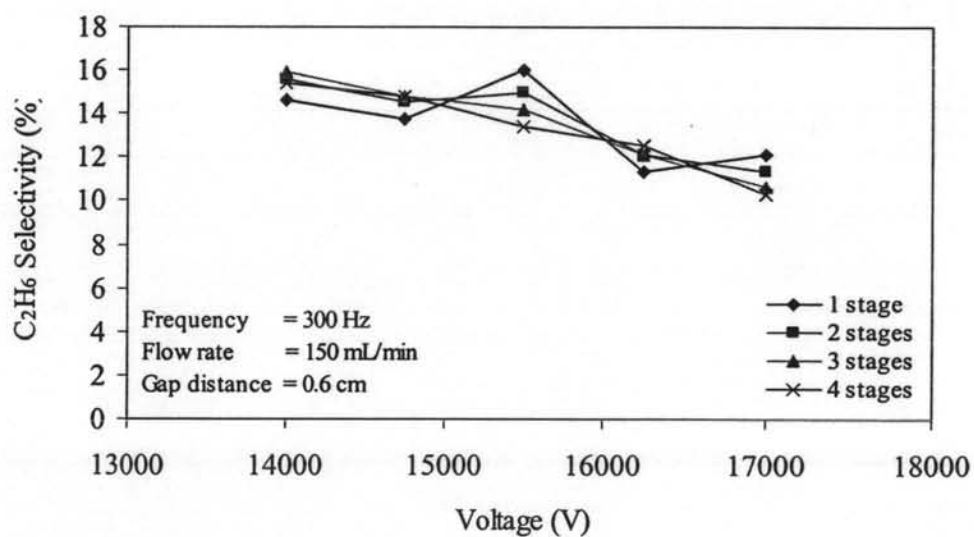


Figure 4.32 Effect of applied voltage on C<sub>2</sub>H<sub>6</sub> selectivity at different stage numbers of the plasma system.

#### 4.4.3 Effect on Power Consumption

Figure 4.33 shows the effect of applied voltage on the power consumption increased. With increasing applied voltage or stage, the power consumption increased until up to 14,750 then the power consumption decreased. In this studied system, 17,000 V was selected for next experiments since this voltage of 17,000 V gave high  $\text{CH}_4$  and  $\text{CO}_2$  conversions as well as the lowest power consumption.

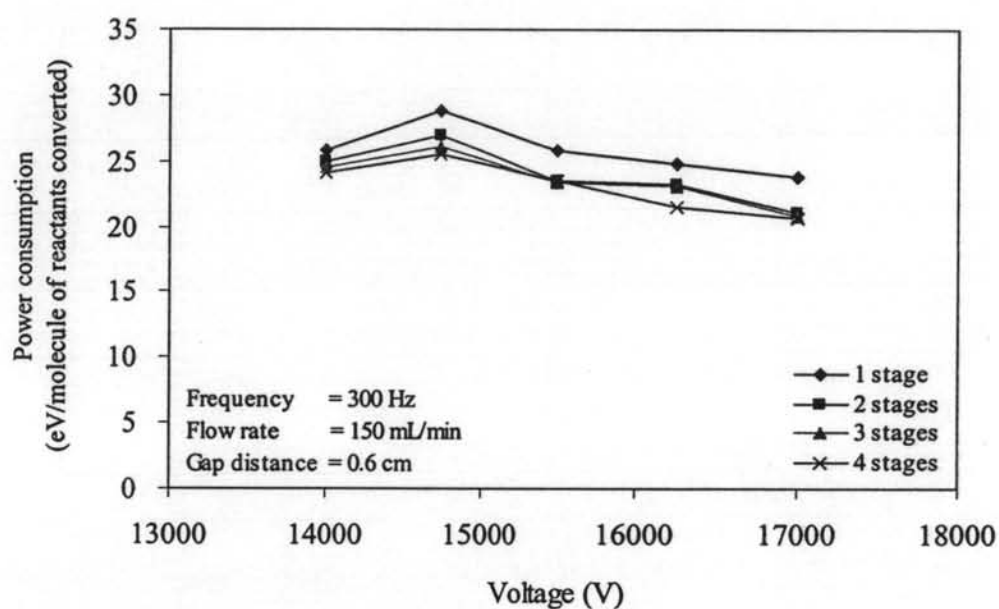


Figure 4.33 Effect of applied voltage on power consumption at different stage numbers of the plasma system.

## 4.5 Effect of Gap Distance

### 4.5.1 Effect on Methane and Carbon Dioxide Conversions

Figures 4.34 and 4.35 show the results of  $\text{CH}_4$  and  $\text{CO}_2$  conversions, respectively. It is interesting note that a maximum gap distance of 0.8 cm could generate plasma under studied system. However, the system could not be operated at a gap distance below 0.6 cm because of the spark was take place in the plasma zone. The  $\text{CH}_4$  and  $\text{CO}_2$  conversions increased with increasing the gap distance similar to that of the stage number. The increase in the gap distance or the stage number simply increases the reaction volume or increasing residence time. As a result, these is more possibility for electrons to collide with  $\text{CH}_4$  and  $\text{CO}_2$  molecules causing higher conversions of both reactants.

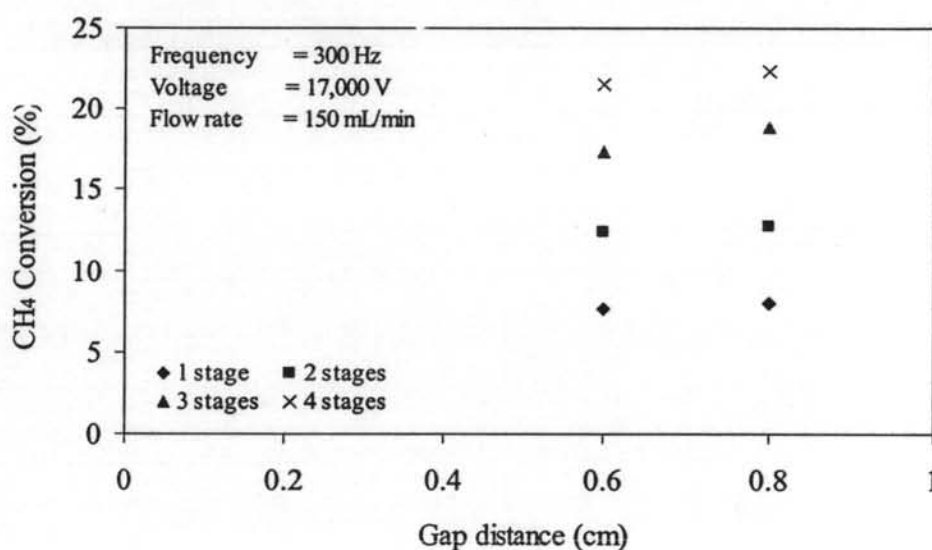
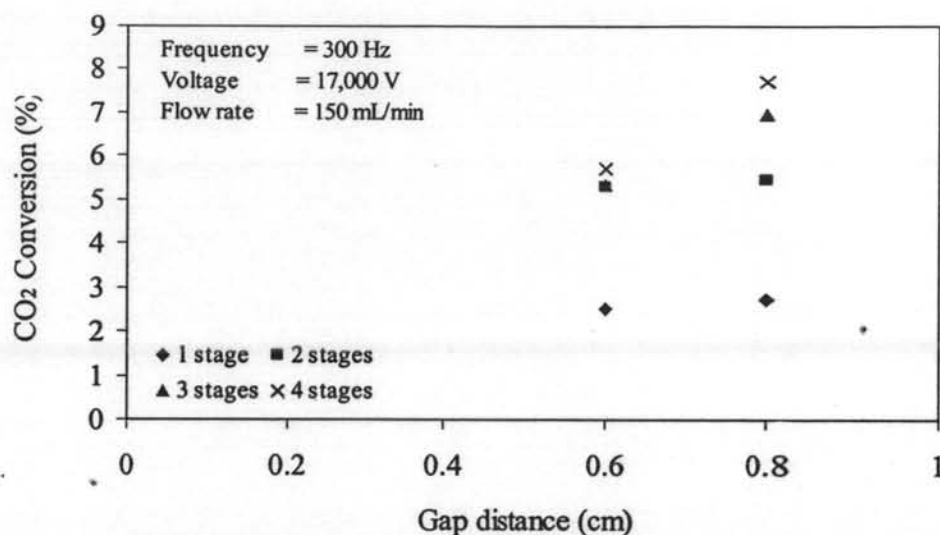


Figure 4.34 Effect of gap distance on  $\text{CH}_4$  conversion at different stage numbers of the plasma system.

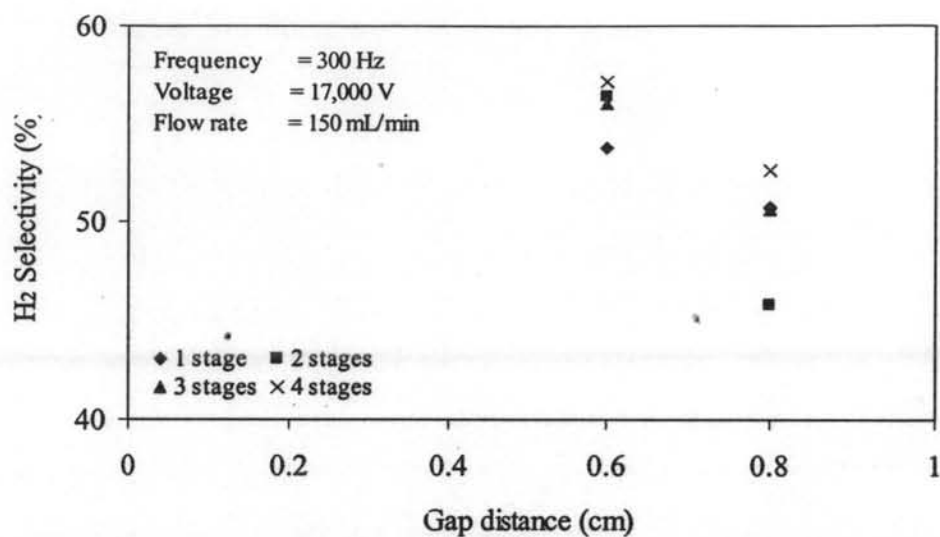


**Figure 4.35** Effect of gap distance on CO<sub>2</sub> conversion at different stage numbers of the plasma system.

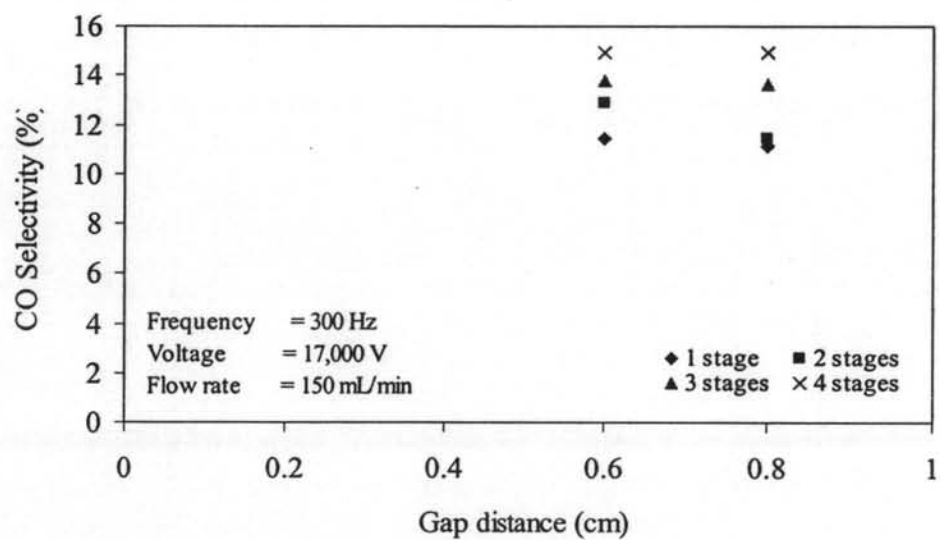
#### 4.5.2 Effect on Product Selectivities

The effect of gap distance on product selectivities are shown in Figures 4.36 to 4.40. With increasing the gap distance, the selectivities of H<sub>2</sub>, CO, C<sub>2</sub>H<sub>2</sub> and C<sub>2</sub>H<sub>4</sub> decreased while the opposite trend was observed for the selectivities of C<sub>2</sub>H<sub>6</sub>. The explanation is that H<sub>2</sub>, CO and C<sub>2</sub>H<sub>2</sub> are the primary products. When either the gap distance or the stage increases, the retention time simply increases leading to higher probability of electrons reacting with the CH<sub>4</sub> molecules to produce primary products. The main composition of biogas is CH<sub>4</sub> that could be produced C<sub>2</sub>H<sub>6</sub> more than synthesis gas.

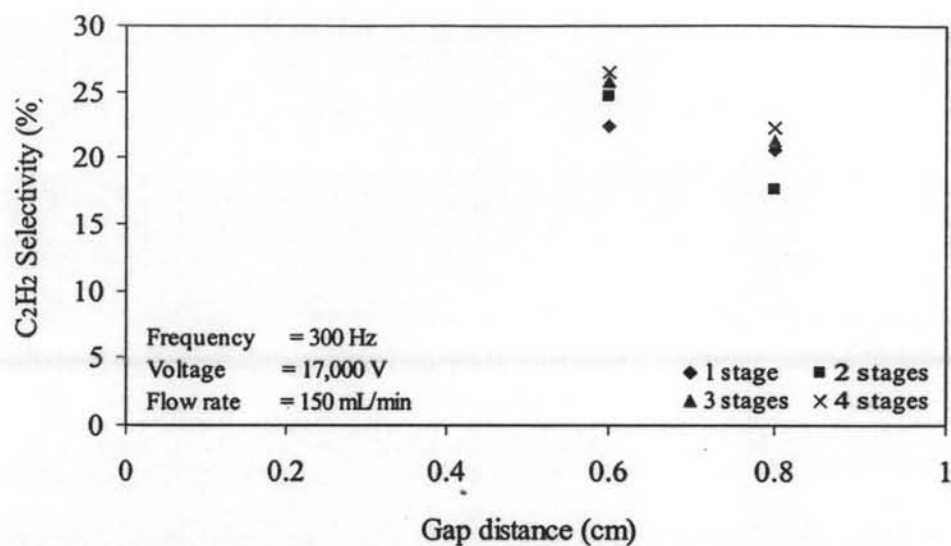




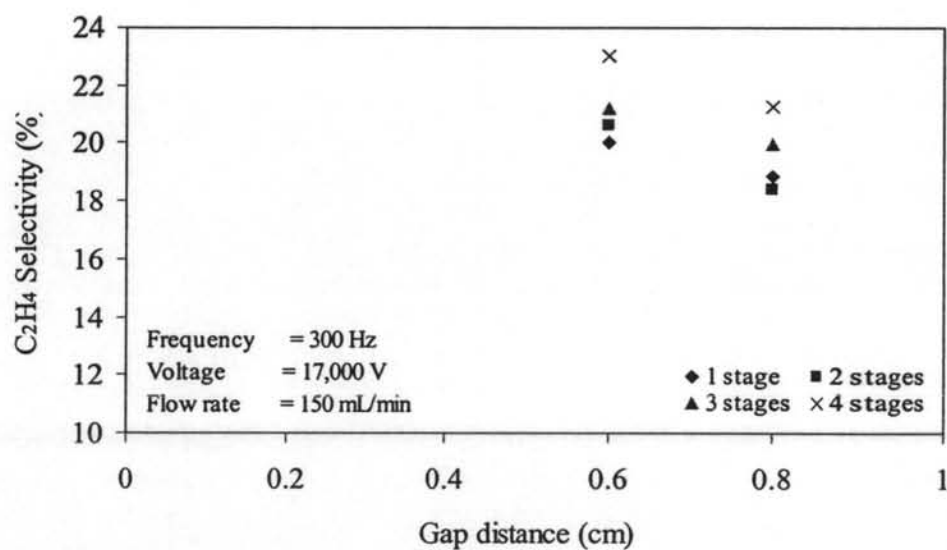
**Figure 4.36** Effect of gap distance on H<sub>2</sub> selectivity at different stage numbers of the plasma system.



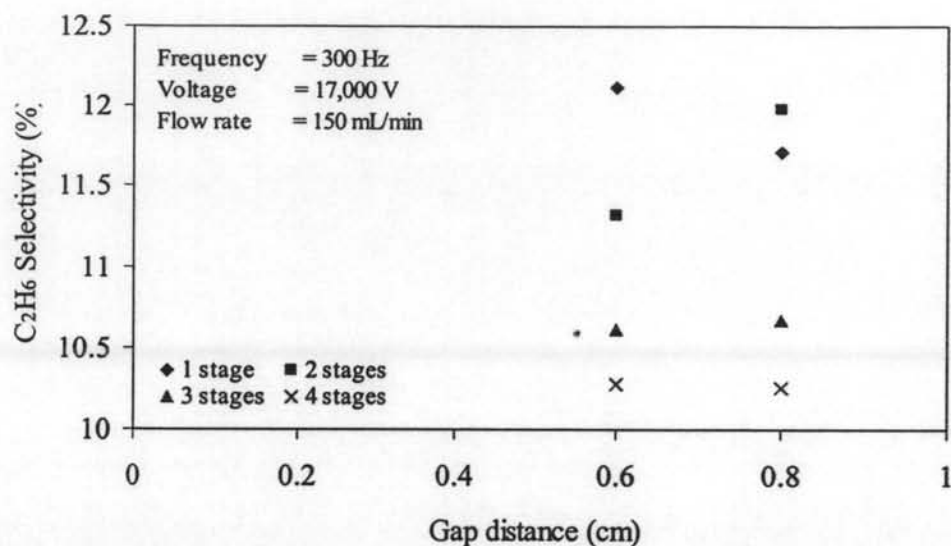
**Figure 4.37** Effect of gap distance on CO selectivity at different stage numbers of the plasma system.



**Figure 4.38** Effect of gap distance on  $C_2H_2$  selectivity at different stage numbers of the plasma system.



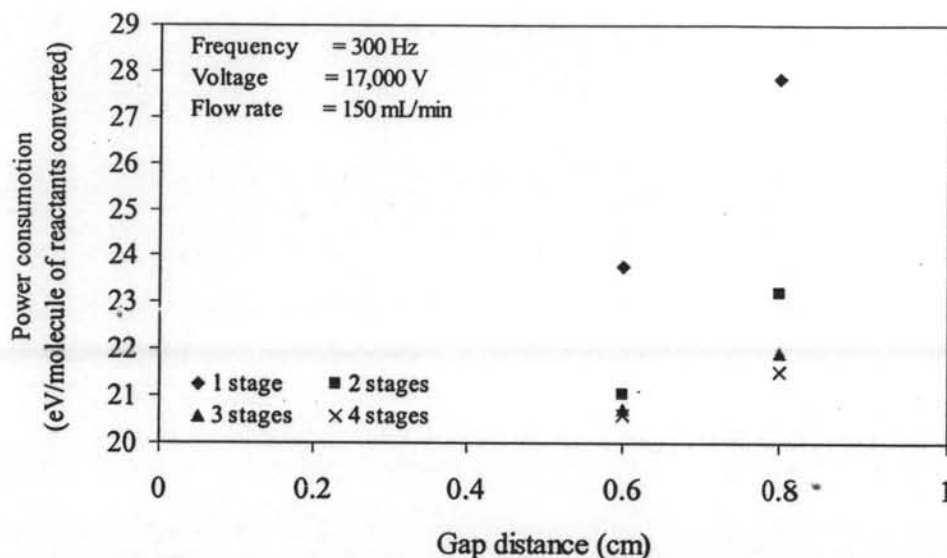
**Figure 4.39** Effect of gap distance on  $C_2H_4$  selectivity at different stage numbers of the plasma system.



**Figure 4.40** Effect of gap distance on  $C_2H_6$  selectivity at different stage numbers of the plasma system.

#### 4.5.3 Effect on Power Consumption

The power consumption increases with increasing gap distance and stage number as shown in Figure 4.41. It indicates that the energy distribution of electrons is improved in a higher gap distance as a result of increasing residence time. The explanation for a significant increase in the power consumption with increasing gap distance is that a number of  $CH_4$  and  $CO_2$  molecule available becomes less and less with respect to lower gap distance. As a result, a gap distance of 0.6 cm was selected to operate the system to determine next parameters.



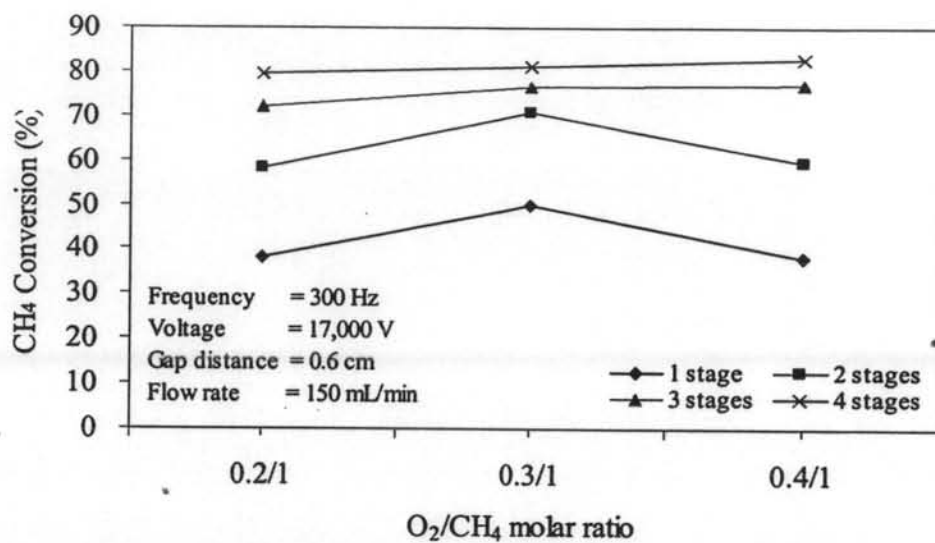
**Figure 4.41** Effect of gap distance on power consumption at different stage numbers of the plasma system.

## 4.6 Effect of Oxygen

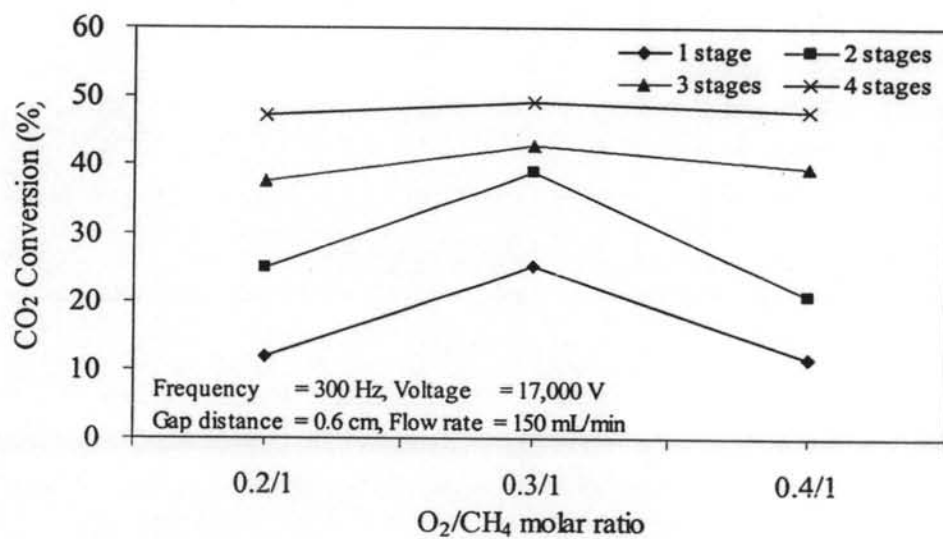
### 4.6.1 Effect on Methane and Carbon Dioxide Conversions

The effects of oxygen concentration on  $\text{CH}_4$ ,  $\text{CO}_2$  and  $\text{O}_2$  conversion are shown in Figures 4.42 to 4.44, respectively. Air was used instead of pure  $\text{O}_2$  in the feed gas since it can reduce investment and operating cost. The Air/Biogas molar ratio was varied from 0.19/1 to 0.38/1 or the  $\text{O}_2/\text{CH}_4$  molar ratio was varied from 0.2/1 to 0.4/1. The conversion of  $\text{CH}_4$ ,  $\text{CO}_2$  and  $\text{O}_2$  increased with increasing  $\text{O}_2$  concentration. The explanation is that a higher amount of  $\text{O}_2$ , the probability of  $\text{CH}_4$  and  $\text{CO}_2$  colliding with the  $\text{O}_2$  molecules increases since there is high  $\text{O}_2$  available. Therefore, it is more chance of methane molecules being activated resulting in higher  $\text{CH}_4$  conversion,  $\text{CO}_2$  conversion and  $\text{O}_2$  conversion.

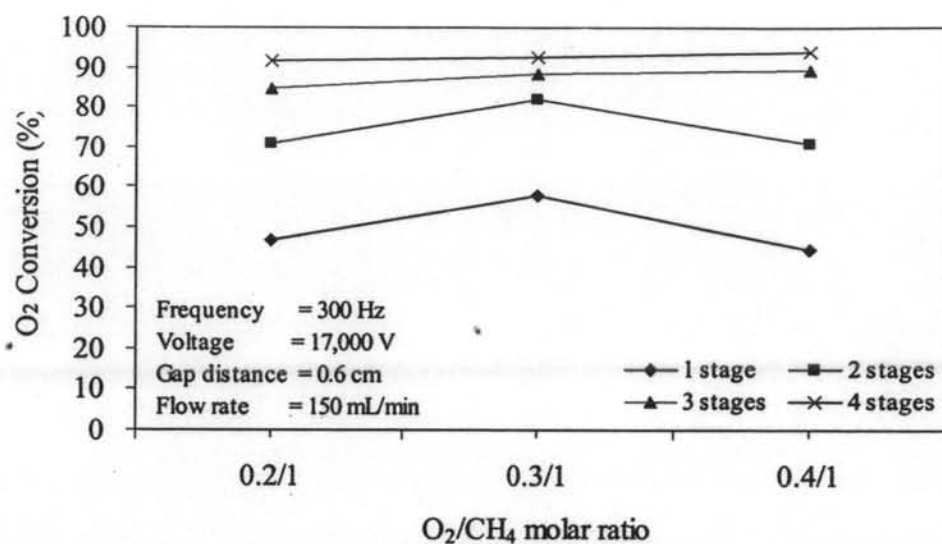
At 1 and 2 stages, the  $\text{CH}_4$ ,  $\text{CO}_2$  and  $\text{O}_2$  conversions decreased when the  $\text{O}_2/\text{CH}_4$  molar ratio is higher than 0.3/1 due to less residence time to convert high amount of  $\text{O}_2$ .



**Figure 4.42** Effect of oxygen on CH<sub>4</sub> conversion at different stage numbers of the plasma system.



**Figure 4.43** Effect of oxygen on CO<sub>2</sub> conversion at different stage numbers of the plasma system.



**Figure 4.44** Effect of oxygen on O<sub>2</sub> conversion at different stage numbers of the plasma system.

#### 4.6.2 Effect on Product Selectivities

Figures 4.45 to 4.48 show the effect of O<sub>2</sub> concentration on the selectivities of H<sub>2</sub>, CO, C<sub>2</sub>H<sub>2</sub> and C<sub>2</sub>H<sub>4</sub>, respectively. When the O<sub>2</sub>/CH<sub>4</sub> molar ratio increased the CO selectivity increased while H<sub>2</sub>, C<sub>2</sub>H<sub>2</sub> and C<sub>2</sub>H<sub>4</sub> decreased. The reason is that increasing O<sub>2</sub>/CH<sub>4</sub> molar ratio results in a lower opportunity of an active methane species reacting with another one resulting in lower dimerization while the system has more oxygen available to form CO. For any given O<sub>2</sub>/CH<sub>4</sub> molar ratio, the selectivities of C<sub>2</sub>H<sub>4</sub> and C<sub>2</sub>H<sub>2</sub> decreased but the selectivity of CO and H<sub>2</sub> increased with increasing stage number. When the stage number is increased corresponding to increasing the residence time, C<sub>2</sub>H<sub>4</sub> is hydrogenated to form C<sub>2</sub>H<sub>2</sub> and C<sub>2</sub>H<sub>2</sub> is further oxidized to form CO. In addition, Ethane was not found because it is the primary product that can be converted to ethylene and acetylene.

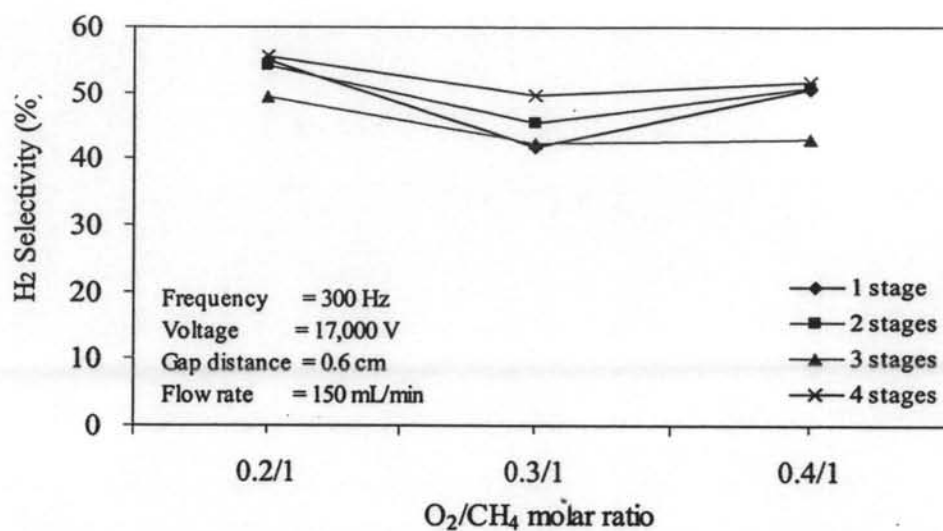


Figure 4.45 Effect of oxygen on H<sub>2</sub> selectivity at different stage numbers of the plasma system.

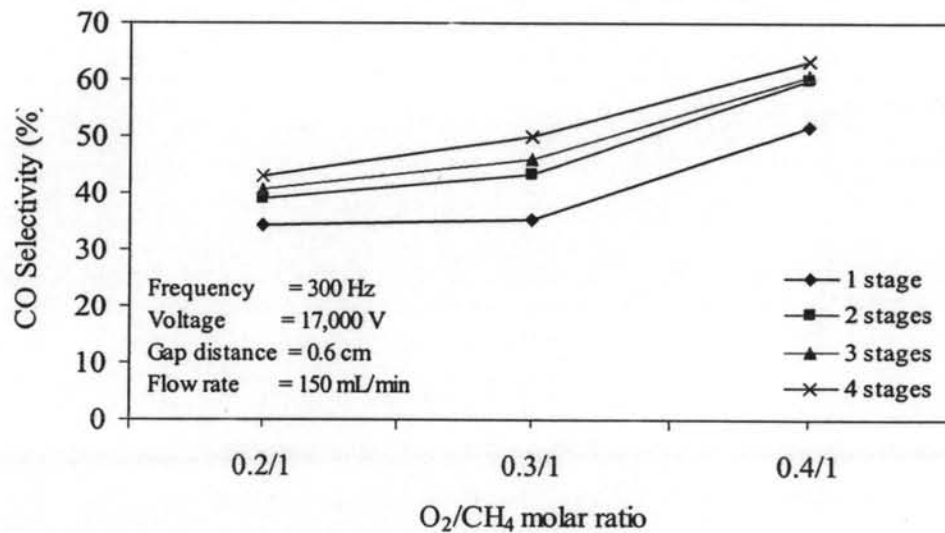
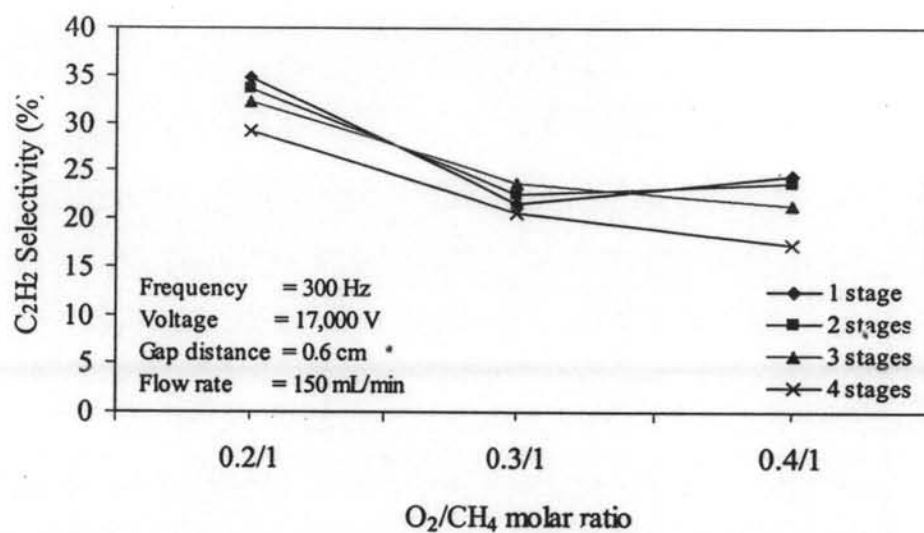
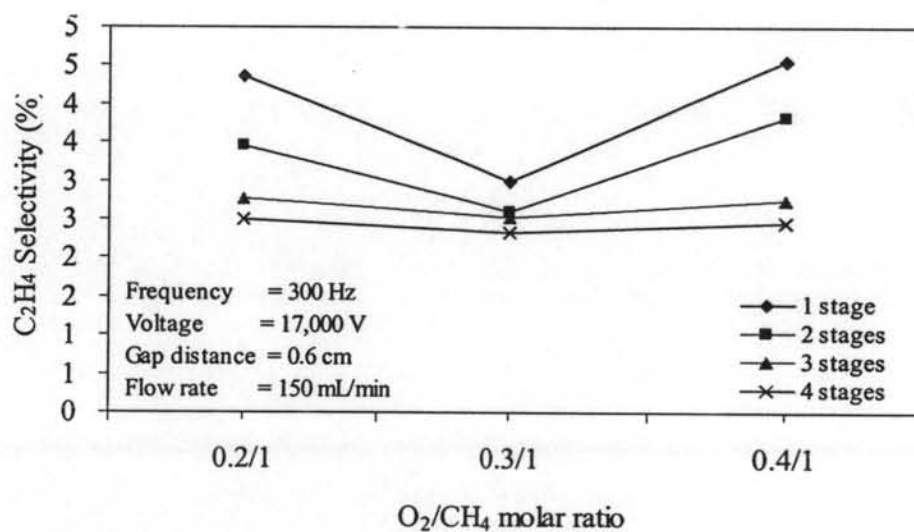


Figure 4.46 Effect of oxygen on CO selectivity at different stage numbers of the plasma system.



**Figure 4.47** Effect of oxygen on C<sub>2</sub>H<sub>2</sub> selectivity at different stage numbers of the plasma system.



**Figure 4.48** Effect of oxygen on C<sub>2</sub>H<sub>4</sub> selectivity at different stage numbers of the plasma system.



#### 4.6.3 Effect on Power Consumption

Figure 4.49 shows the effect of oxygen concentration on power consumption. The power consumption decreased with increasing  $O_2/CH_4$  molar ratio and then increased. Increasing  $O_2/CH_4$  molar ratio results in easier activation of  $CH_4$  and  $CO_2$  molecules because the ionization energy of oxygen is lower than ionization of  $CH_4$  and  $CO_2$ . For  $O_2/CH_4$  molar ratio of 0.4/1, the power consumption increased due to  $CH_4$ ,  $CO_2$  and  $O_2$  conversion decreased.

The  $O_2/CH_4$  molar ratio of 0.3/1 should be selected for biogas reforming because it had high  $CH_4$ ,  $CO_2$  and  $O_2$  conversion and lower power consumption.

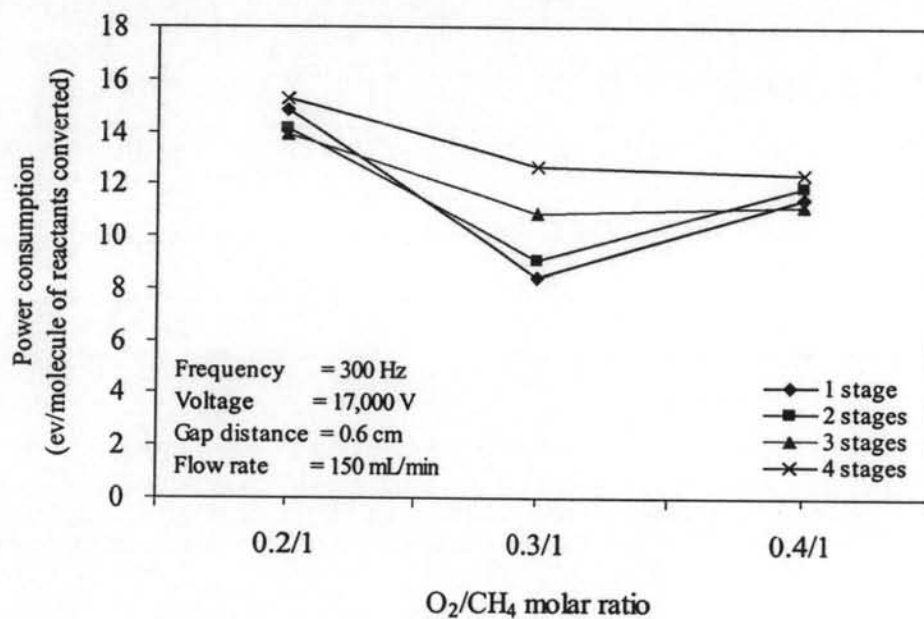


Figure 4.49 Effect of oxygen on power consumption at different stage numbers of the plasma system.