CHAPTER IV
RESULTS AND DISCUSSION

4.1 Effect of Molar Ratio

4.1.1 Effect on Methane and Oxygen Conversion

The effects of CH_4/O_2 molar ratio on the methane and oxygen conversion are shown in Figures 4.1 and 4.2, respectively. The CH_4/O_2 molar ratio was varied from 2/1 to 5/1. The conversion of CH_4 and O_2 decreased with increasing CH_4/O_2 molar ratio. The highest CH_4 and O_2 conversion were at a CH_4/O_2 molar ratio 2/1. The explanation is that a higher CH_4/O_2 molar ratio, the probability of methane colliding with the O_2 molecules decreases since there is less oxygen available. Therefore, it is less chance of methane molecules being activated resulting in lower methane conversion and oxygen conversion. For any given CH_4/O_2 molar ratio, the CH_4 and O_2 conversions increased substantially with increasing stage number of reactor. This is because the residence time increases as an increase in the stage number.

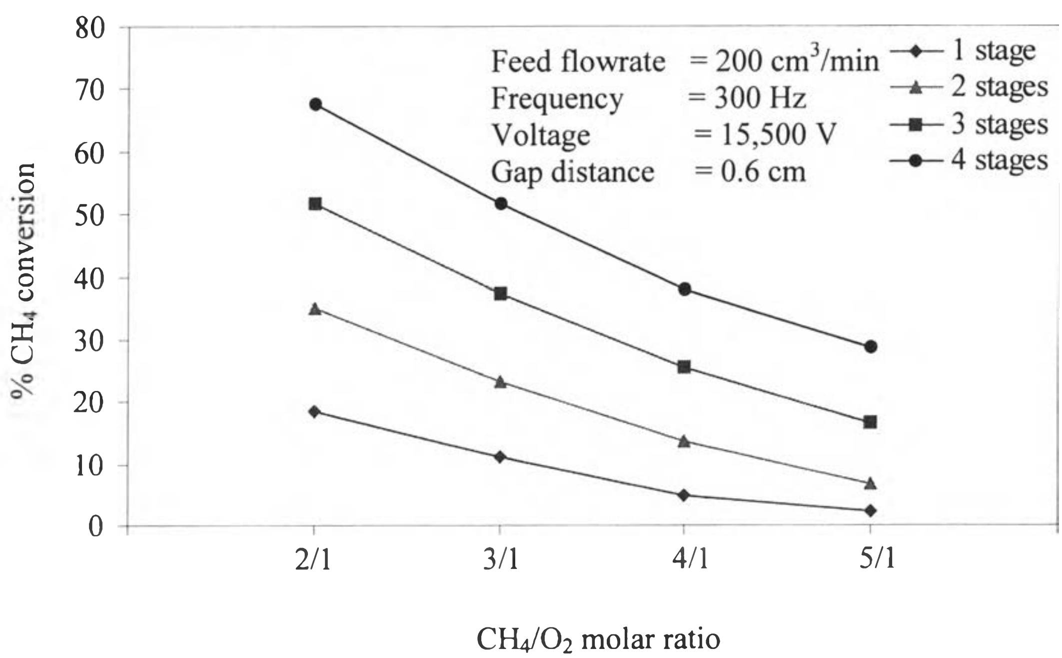

Figure 4.1 Effect of CH_4/O_2 molar ratio on CH_4 conversion at different stage numbers of the plasma system.

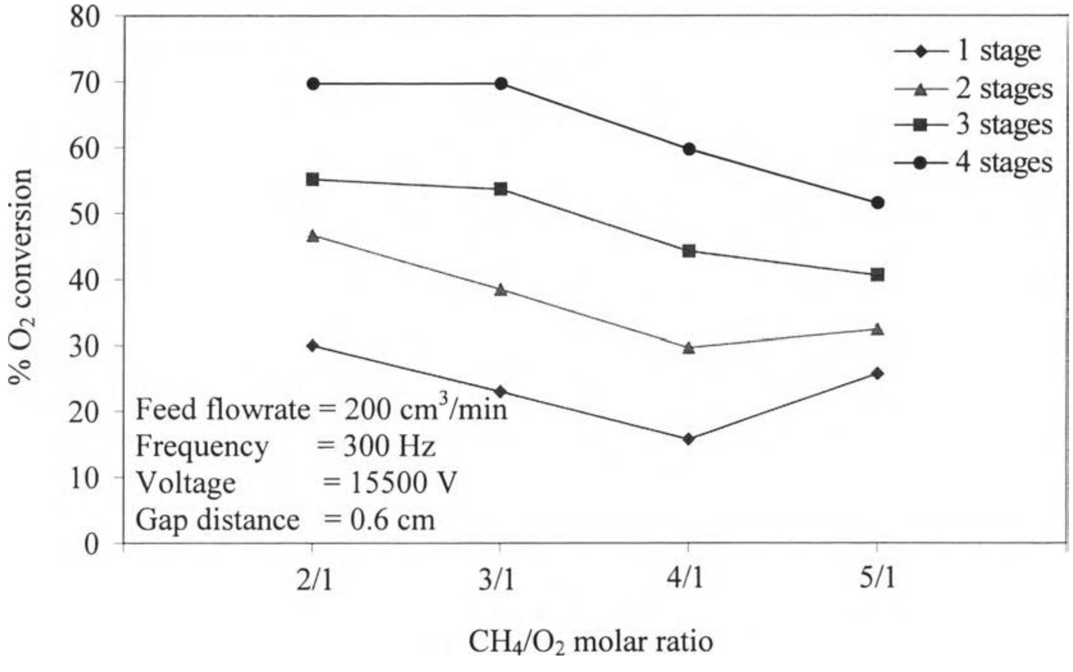

Figure 4.2 Effect of CH_4/O_2 molar ratio on O_2 conversion of at different stage numbers of the plasma system.

4.1.2 Effect on Product Selectivities

Figures 4.3 to 4.8 show the effect of CH_4/O_2 molar ratio on the selectivities of CO, H_2, CO_2, C_2H_2, C_2H_4 and C_2H_6, respectively. When the CH_4/O_2 molar ratio increased the CO and CO_2 selectivity decreased while H_2, C_2H_2, C_2H_4 and C_2H_6 increased. The reason is that increasing CH_4/O_2 molar ratio results in a higher opportunity of an active methane species reacting with another one resulting in dimerization while the system has less oxygen available to form CO and CO_2. For any given CH_4/O_2 molar ratio, the selectivities of CO, H_2, C_2H_4 and C_2H_6 decreased but the selectivity of CO_2 and C_2H_2 increased with increasing stage number. The results imply that of CO, H_2, C_2H_4 and C_2H_6 are the primary products. When the stage number is increased corresponding to increasing the residence time, CO is further oxidized to form CO_2 and there are more C_2H_6 to be formed from coupling reaction of methane. In addition, the rate of dehydrogenation increases with increasing stage number of reactors.

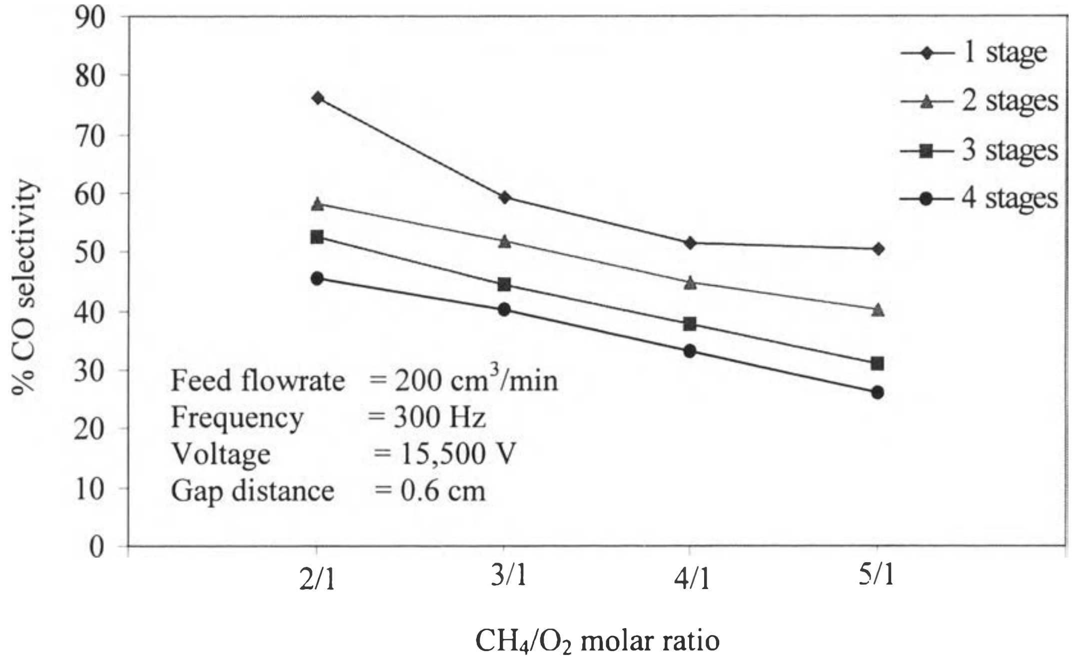

Figure 4.3 Effect of CH_4/O_2 molar ratio on CO selectivity of at different stage numbers of the plasma system.

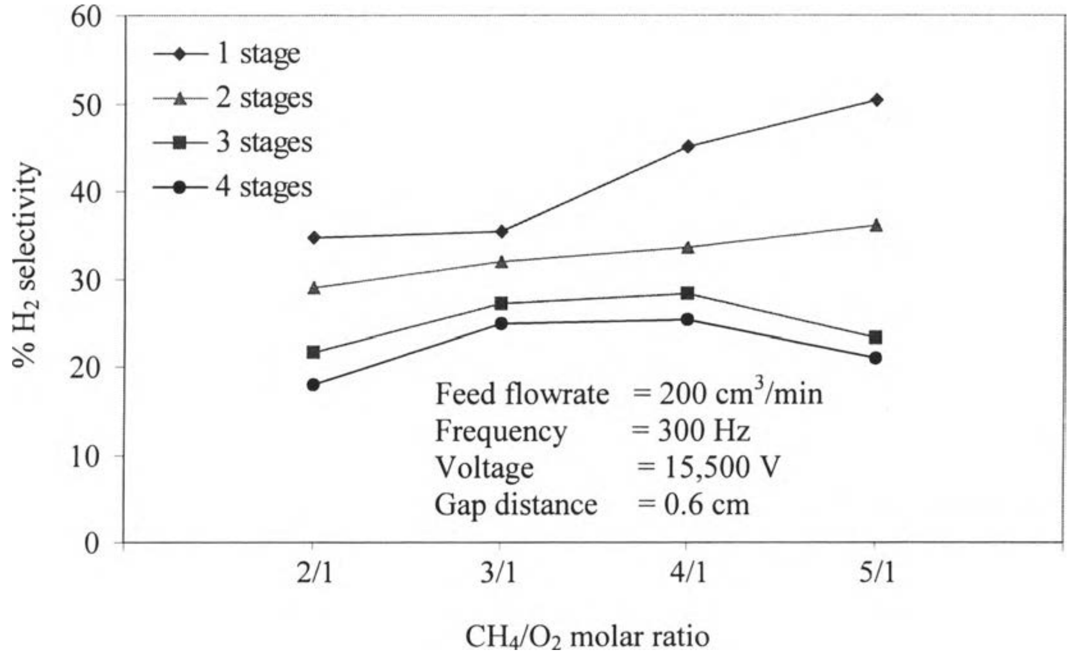

Figure 4.4 Effect of CH_4/O_2 molar ratio on H_2 selectivity at different stage numbers of the plasma system.

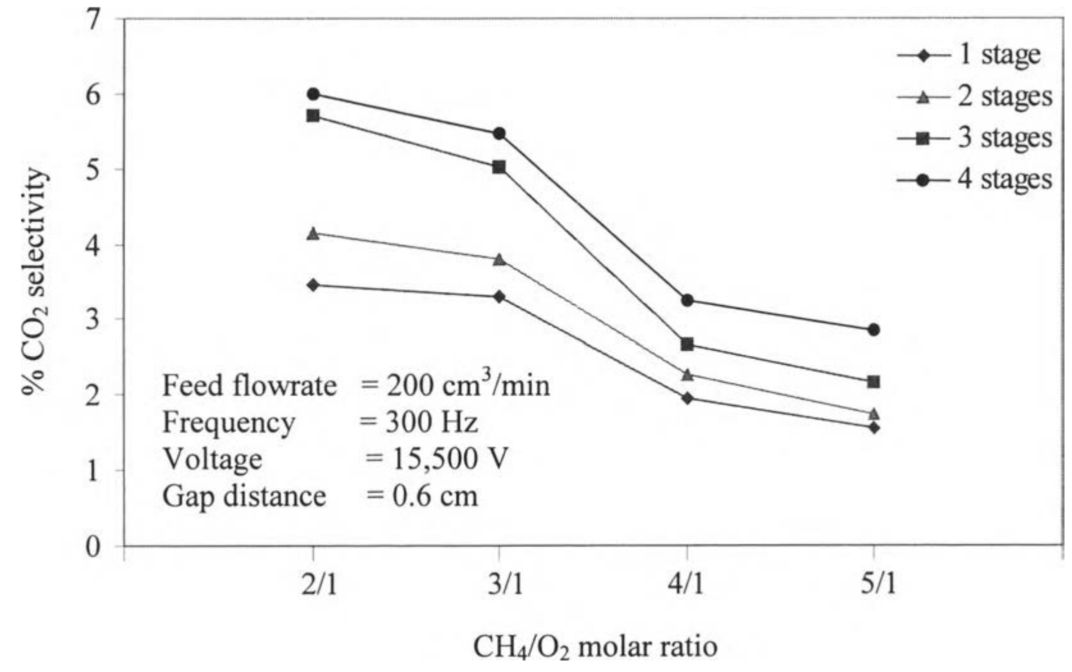

Figure 4.5 Effect of CH_4/O_2 molar ratio on CO_2 selectivity at different stage numbers of the plasma system.

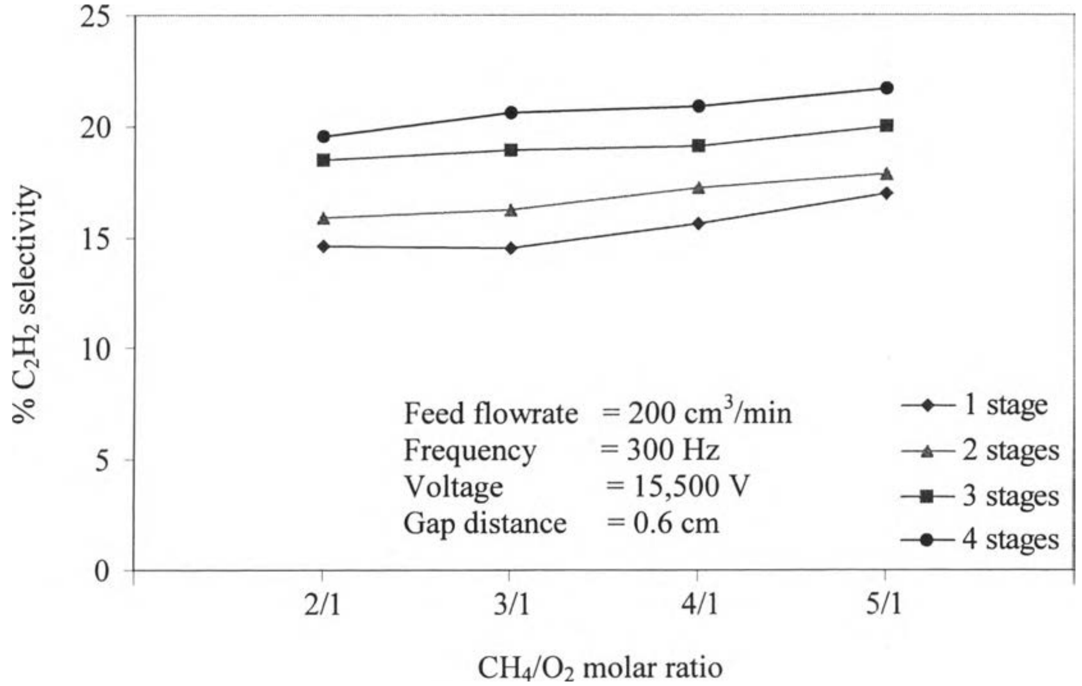

Figure 4.6 Effect of CH_4/O_2 molar ratio on C_2H_2 selectivity at different stage numbers of the plasma system.

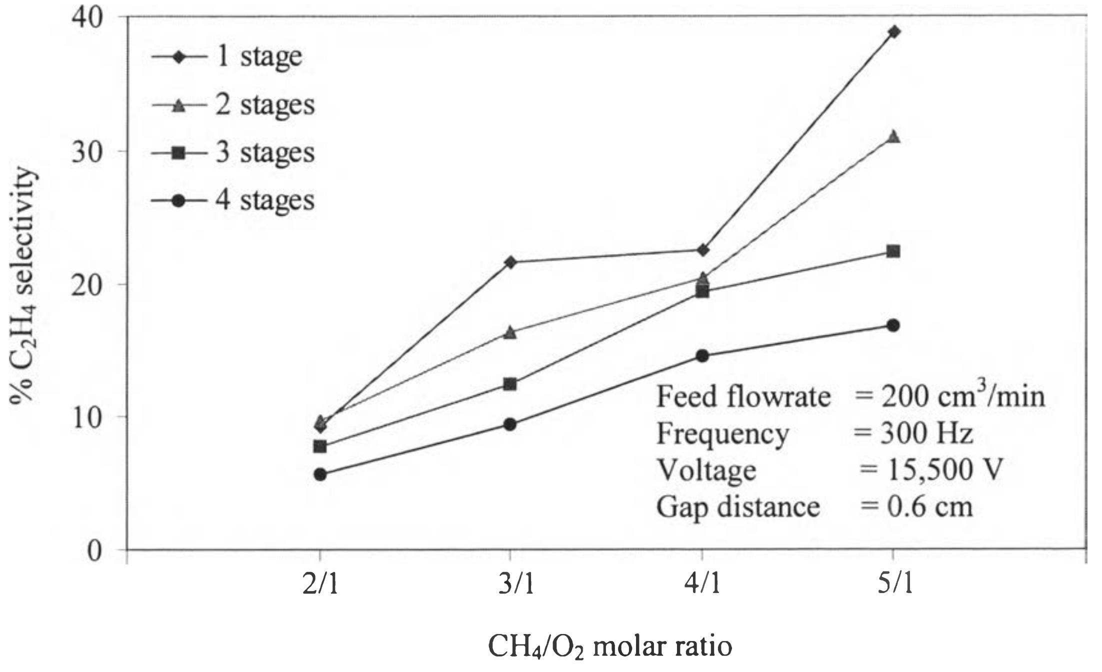

Figure 4.7 Effect of CH_4/O_2 molar ratio on C_2H_4 selectivity at different stage number of the plasma system.

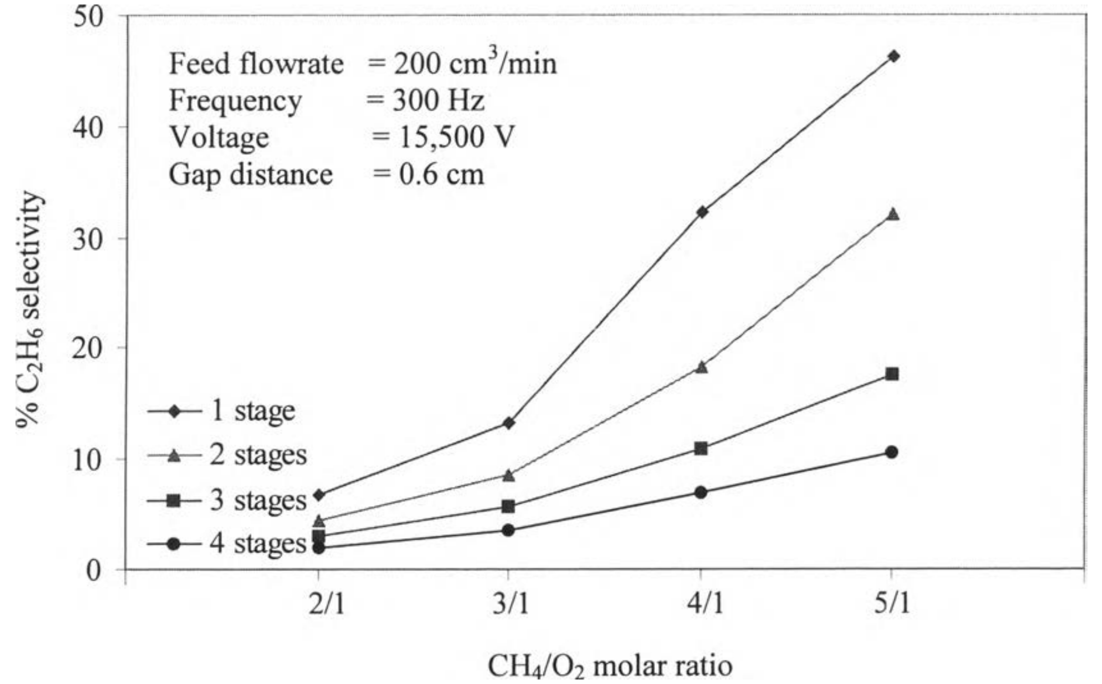

Figure 4.8 Effect of CH_4/O_2 molar ratio on C_2H_6 selectivity at different stage numbers of the plasma system.

4.1.3 Effect on Power Consumption

Figure 4.9 shows the effect of CH_4/O_2 feed molar ratio on power consumption of methane molecules. The power consumption increased with increasing CH_4/O_2 feed molar ratio. Since CH_4/O_2 feed molar ratio decreased result in increasing methane conversion. An optimum power is approximately accomplished when the CH_4/O_2 feed molar ratio is in the range of 3/1 to 5/1 CH_4/O_2 feed molar ratio.

Importantly, the minimum power consumption was found in the range of 3/1 to 5/1 CH_4/O_2 feed molar ratio. But 3/1 CH_4/O_2 in feed was selected for next experiments because it had higher CH_4 and O_2 conversion and lower by-products than other CH_4/O_2 feed mole ratios.

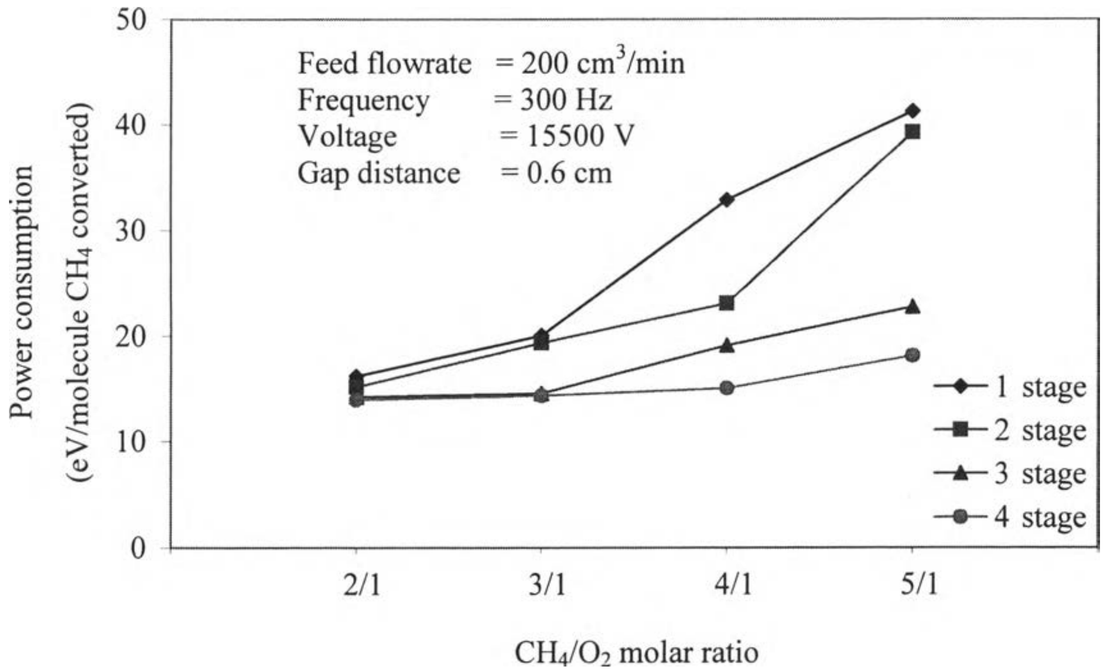

Figure 4.9 Effect of molar ratio on power consumption at different stage numbers of the plasma system.

4.2 Effect of Feed Flowrate

4.2.1 Effect on Methane and Oxygen Conversion

The effects of feed flowrate on CH_4 and O_2 conversions are illustrated in Figures 4.10 and 4.11, respectively. Feed flowrate is one of the most important operating parameters affecting the performance of the plasma system because it directly relates to residence time. Both CH_4 and O_2 conversions decreased with increasing feed flowrate from 50-300 cm^3/min. This result leads to a conclusion that either the feed flowrate increases or the stage number decreases resulting in decreasing residence time. Therefore the contact time of methane and oxygen molecules with electrons decreases leading to lower conversion of both CH_4 and O_2.

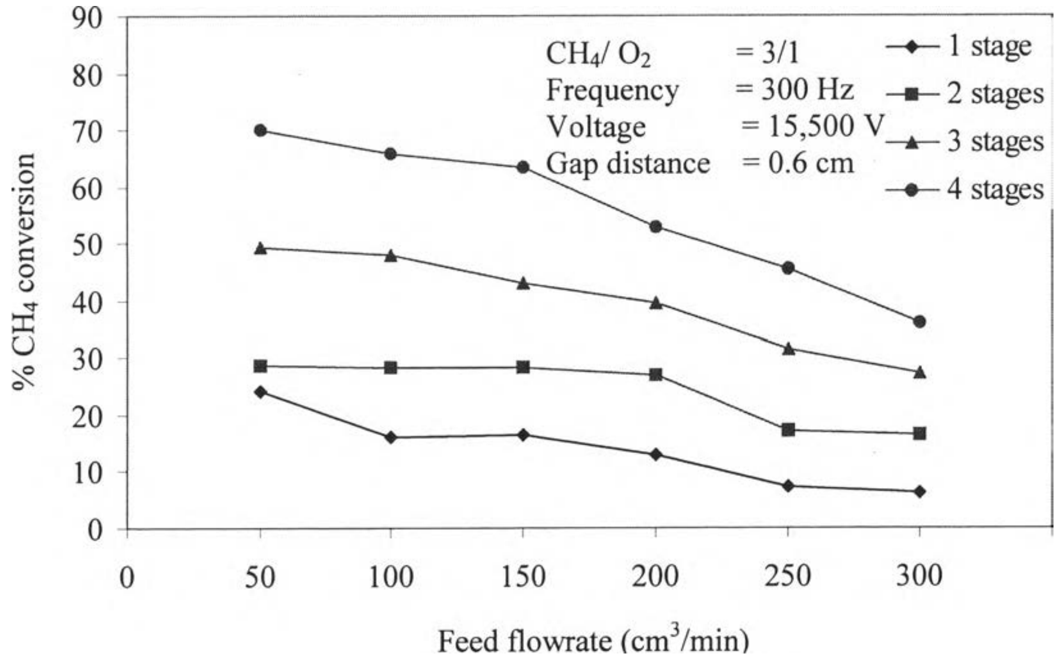

Figure 4.10 Effect of feed flowrate on CH_4 conversion at different stage numbers of the plasma system.

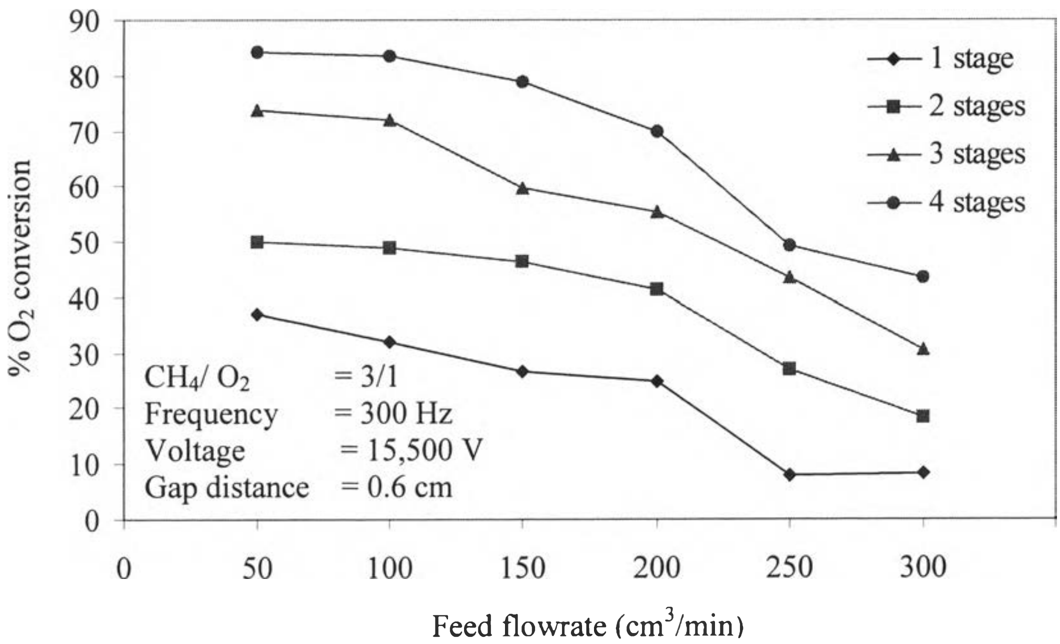

Figure 4.11 Effect of feed flowrate on O_2 conversion at different stage numbers of the plasma system.

4.2.2 Effect on Product Selectivities

Figures 4.12 to 4.17 show the effect of feed flowrate on the selectivities of CO, H_2, CO_2, C_2H_2, C_2H_4 and C_2H_6, respectively. As the feed flowrate increased, the CO selectivity increased, in the contrast the selectivities of H_2, CO_2 and C_2H_2 decreased and the selectivities of C_2H_4 and C_2H_6 slightly decreased. The results can be explained that the feed flowrate was decreased leading to increasing residence time. As a result, more methane molecules will react with oxygen to form more CO and CO_2 as well as other reactions of CO oxidation, the methane coupling and dehydrogenation are also enhanced.

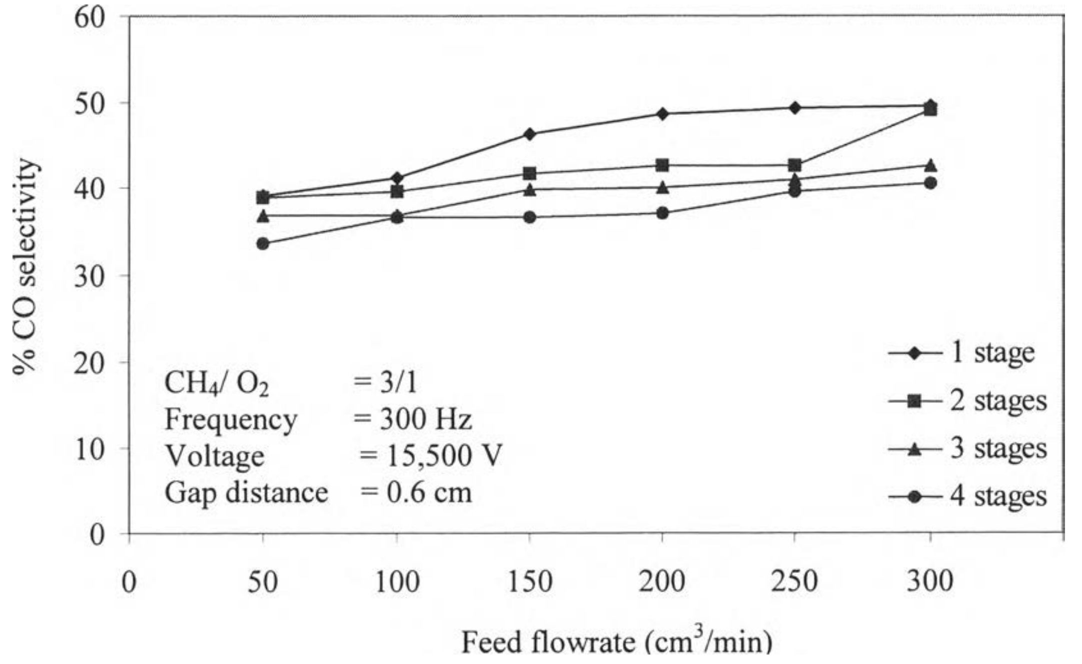

Figure 4.12 Effect of feed flowrate on CO selectivity at different stage numbers of the plasma system.

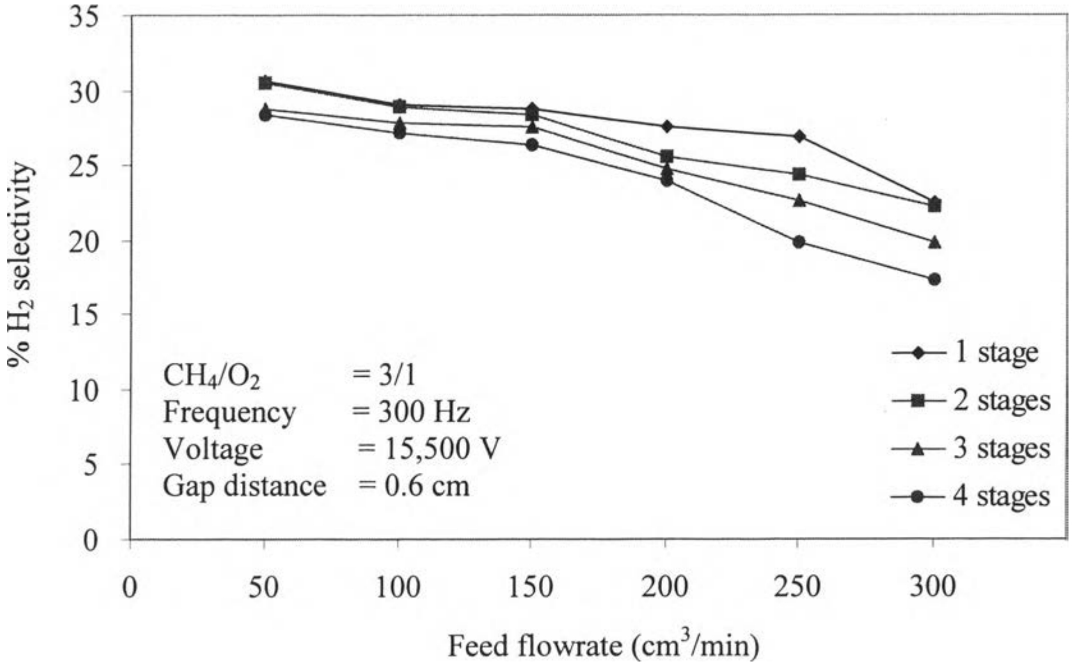

Figure 4.13 Effect of feed flowrate on H_2 selectivity at different stage numbers of the plasma system.

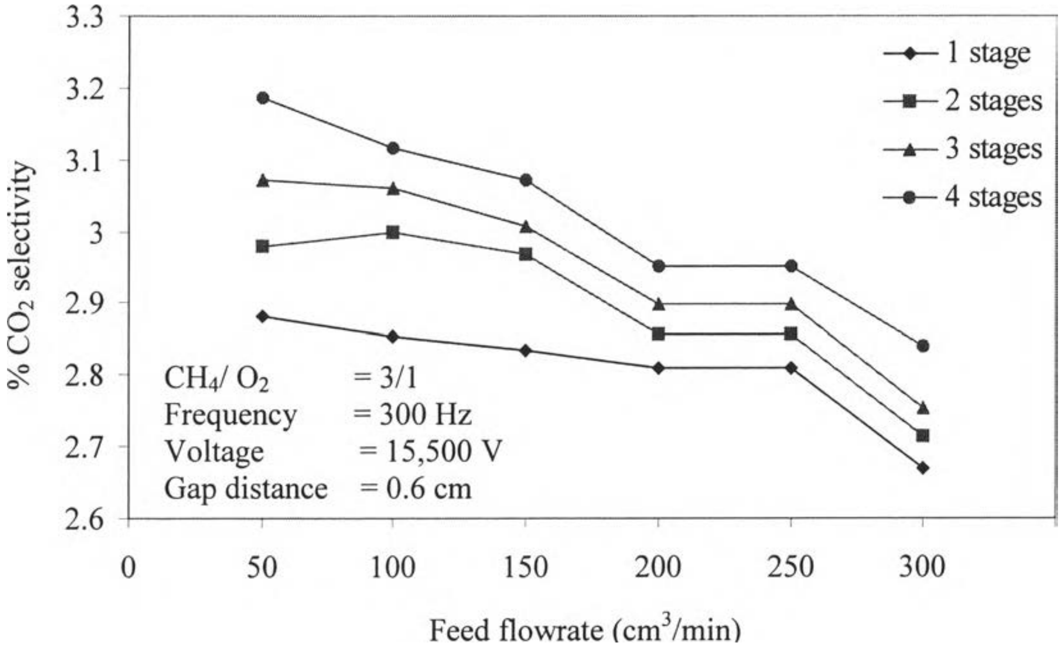

Figure 4.14 Effect of feed flowrate on CO_2 selectivity at difference stage number of the plasma system.

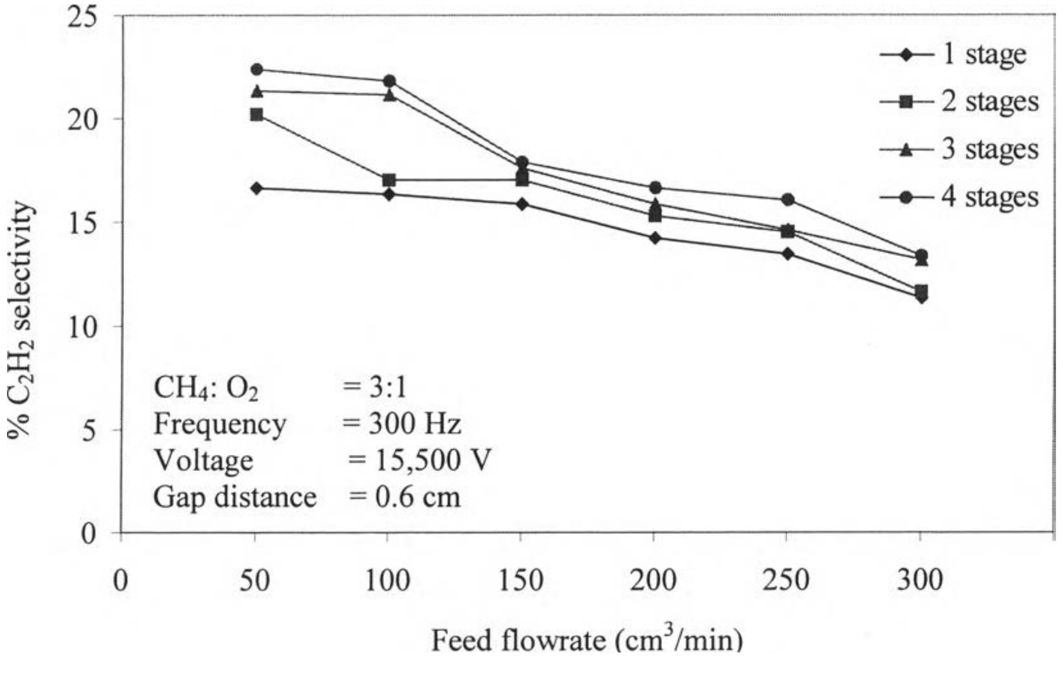

Figure 4.15 Effect of feed flowrate on C_2H_2 selectivity at different stage numbers of the plasma system.

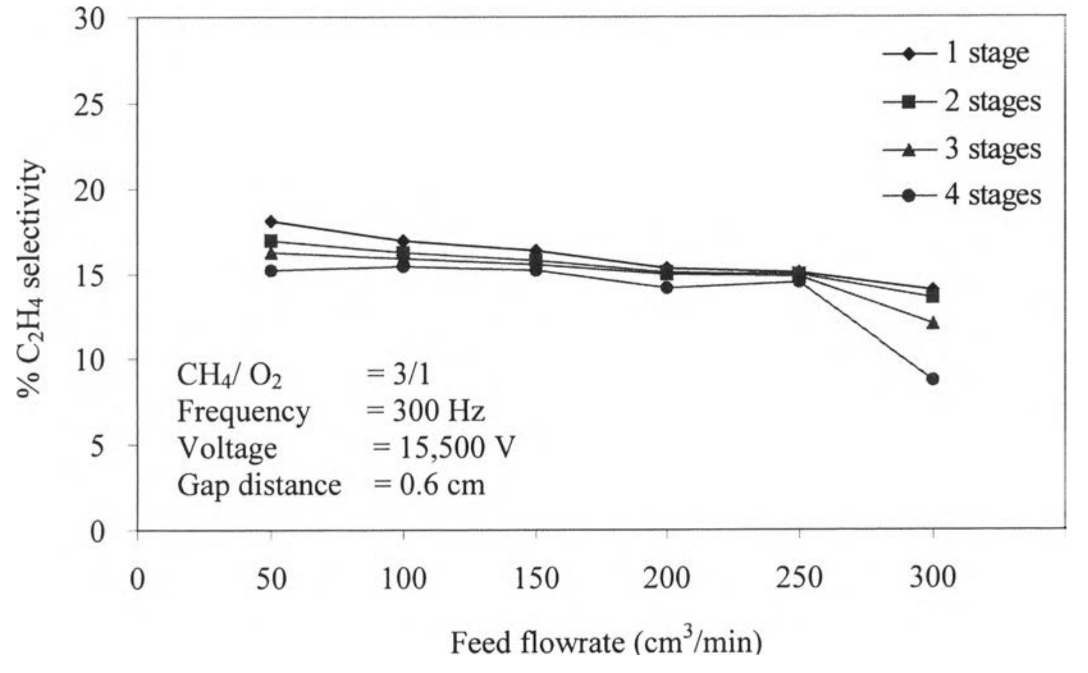

Figure 4.16 Effect of feed flowrate on C_2H_4 selectivity at different stage numbers of the plasma system.

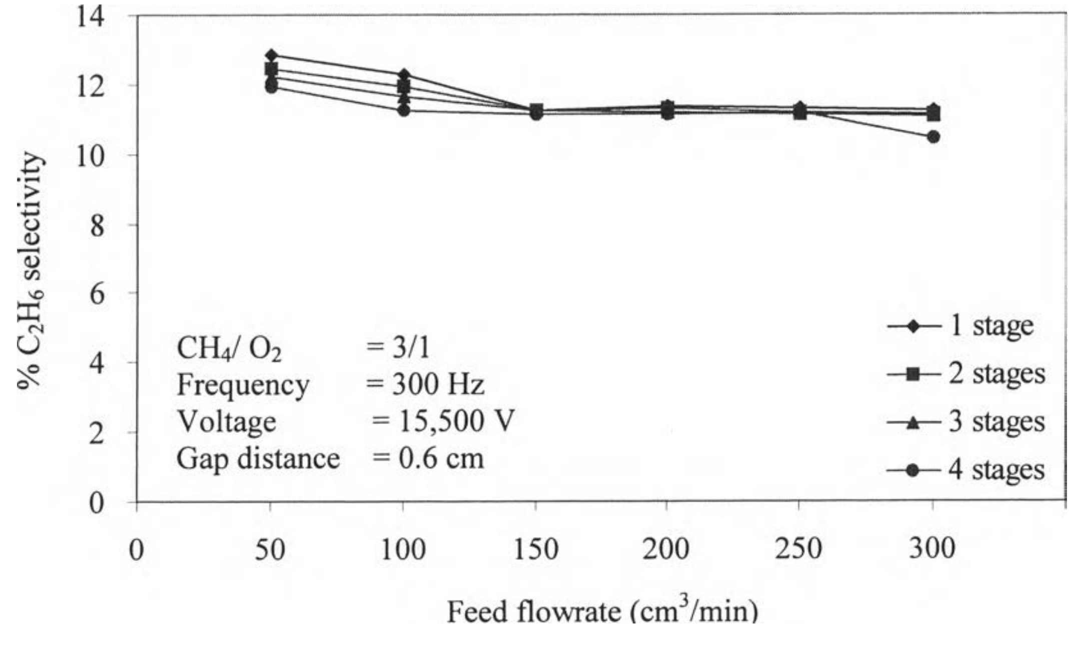

Figure 4.17 Effect of feed flowrate on C_2H_6 selectivity at different stage numbers of the plasma system.

4.2.3 Effect on Power Consumption

As can be seen from Figure 4.18, the minimum power consumption is approximately achieved at the feed flowrate about 150-200 cm^3/min. At a flowrate higher than 200 cm^3/min, power consumption increased remarkly with increasing flowrate since the electrons produced have less propability to collide with O_2 and CH_4 to produce the active species. At a flowrate lower than 150 cm^3/min, power consumption increased with decreasing feed flowrate because of lower probabilities of reacting with lower amounts of reactants available and some of the power may be consumed by secondary reactions. (Supat *et al.*, 2003)

According to the power consumption, a feed flowrate in the range of 150-200 cm^3/min should be considered for operating this plasma reactor system. Even though the system has the lower power consumption at a feed flowrate of 200 cm^3/min, it had a lower methane conversion as well as a lower synthesis gas production as compared to that operated at a feed flowrate of 150 cm^3/min. As a result, a feed flowrate of 150 cm^3/min was selected to operate the system to determine other parameters.

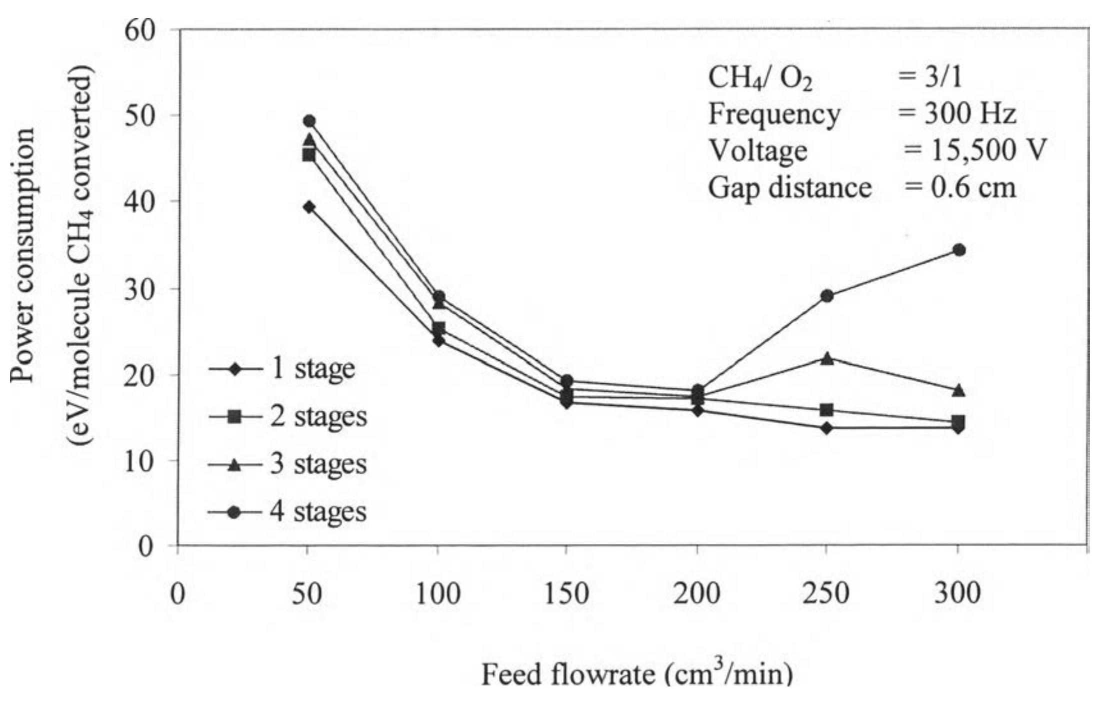

Figure 4.18 Effect of feed flowrate on power consumption at different stage numbers of the plasma system.

4.3 Effect of Residence Time

4.3.1 Effect on Methane and Oxygen Conversions

The effect of residence time on the process performance of the gliding arc system was studied by varying the feed flowrate from 50 to 300 cm^3/min corresponding to residence time of 0.55 to 3.31 seconds. The reaction volume was determined from the observation of the plasma appearance in each reactor which was approximately 3 cm^3 for each reactor. The results of methane and oxygen conversions are shown in Figures 4.19 and 4.20, respectively. The conversions of CH_4 and O_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 reacton zone is obtained resulting in more chance of both CH_4 and O_2 molecules to be contacted with electrons. As a result, both CH_4 and O_2 conversions increase with increasing residence time.

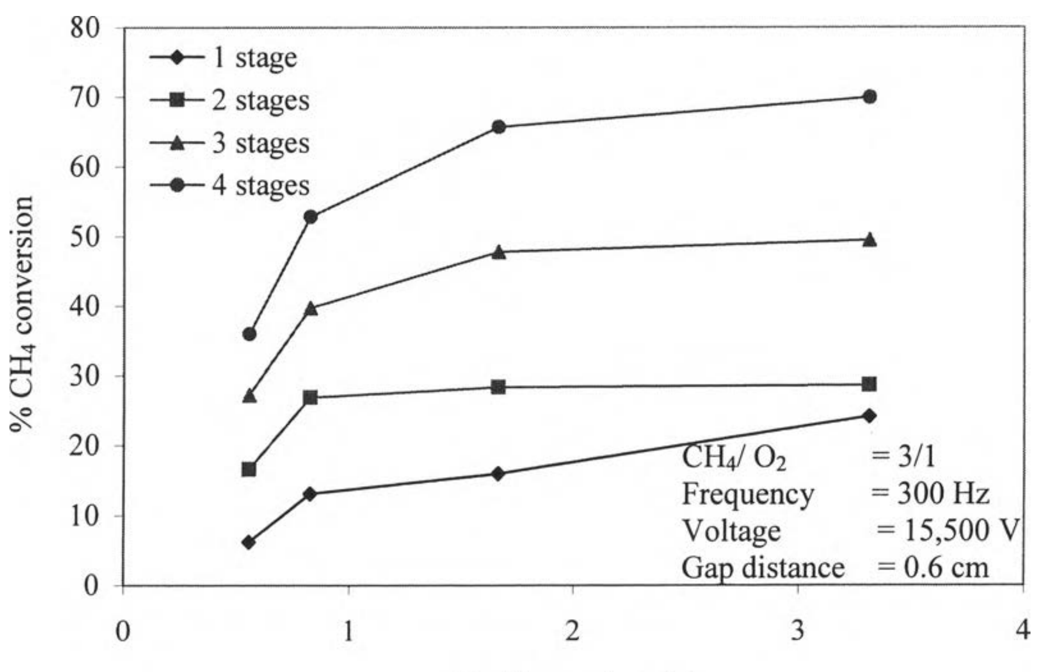

Figure 4.19 Effect of residence time on CH_4 conversion at different stage numbers of the plasma system.

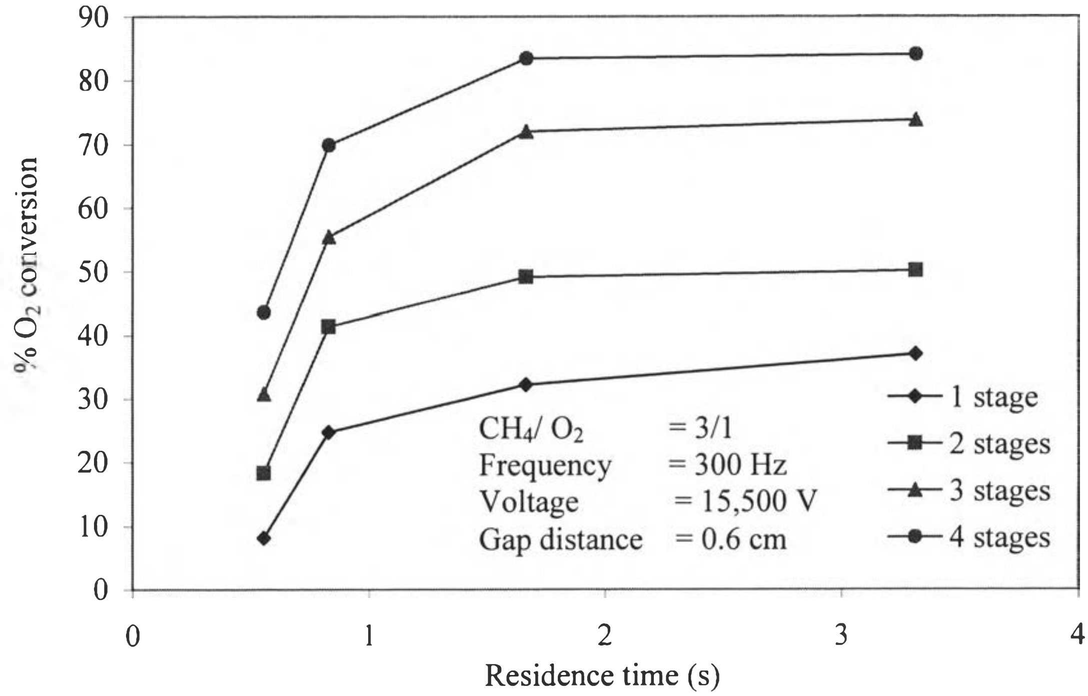

Figure 4.20 Effect of residence time on O_2 conversion at different stage numbers of the plasma system.

4.3.2 Effect on Product Selectivities

Figures 4.21 to 4.26 show the effect of residence time on the product selectivities. The selectivities of CO, H_2, C_2H_4 and C_2H_6 slightly decreased while the selectivities of CO_2, and C_2H_2 slightly increased with increasing residence time. In addition, the stage number of the plasma system has no strong effect on the product selectivities. The explanation for the effect of residence time is the same as for the effect of feed flowrate.

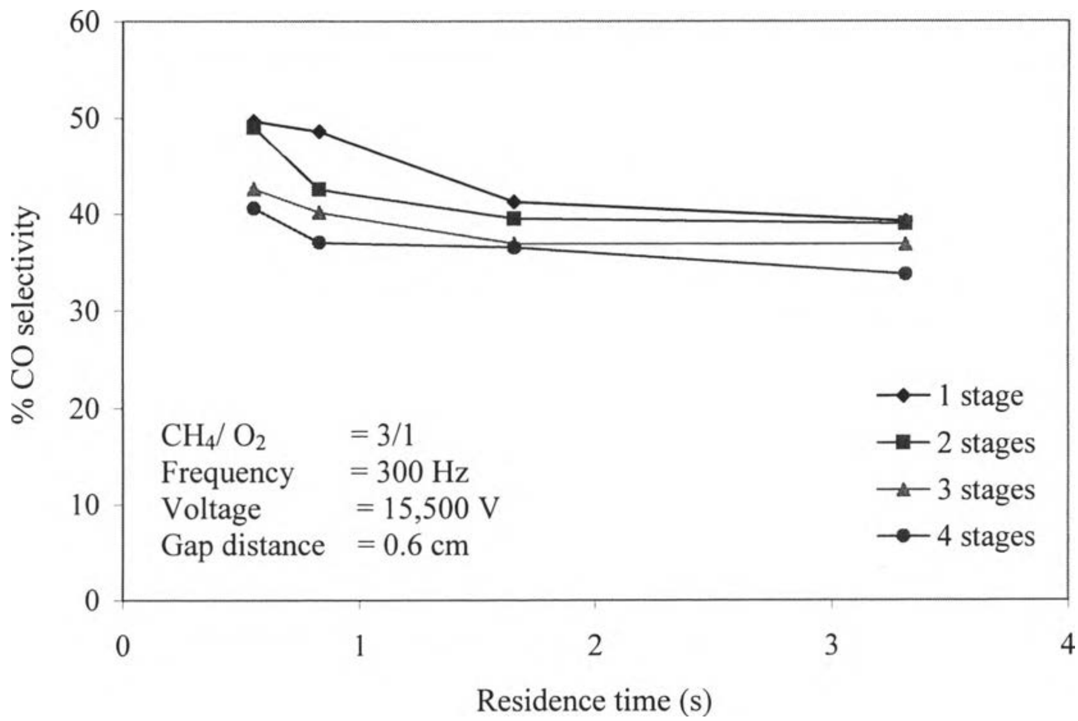

Figure 4.21 Effect of residence time on CO selectivity at different stage numbers of the plasma system.

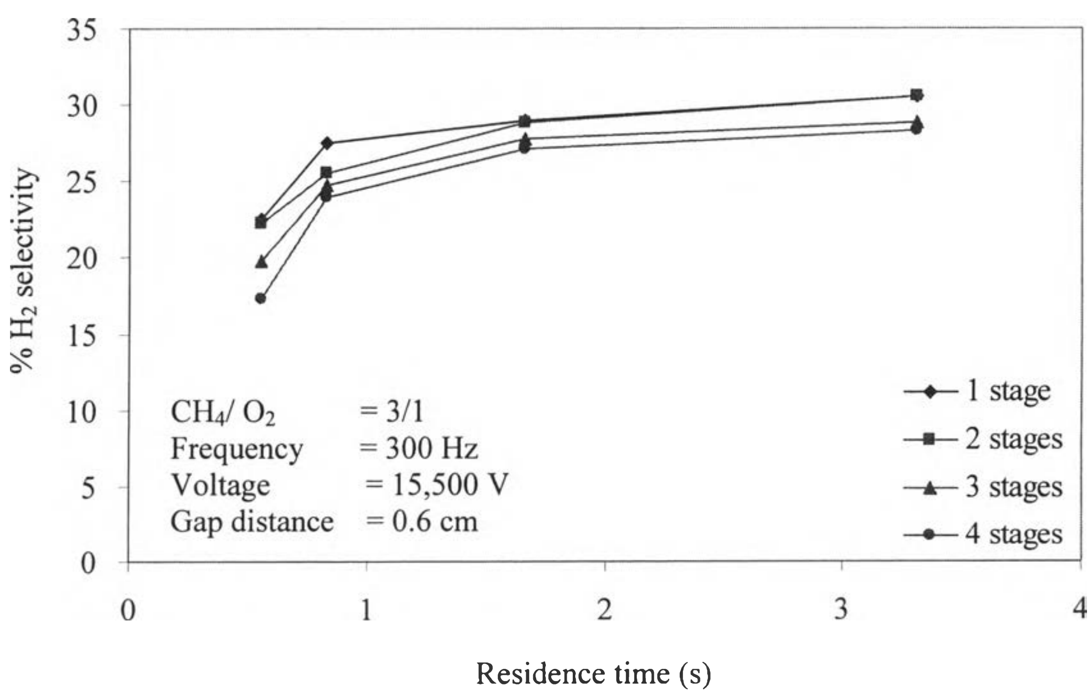

Figure 4.22 Effect of residence time on H_2 selectivity of at different stage numbers of the plasma system.

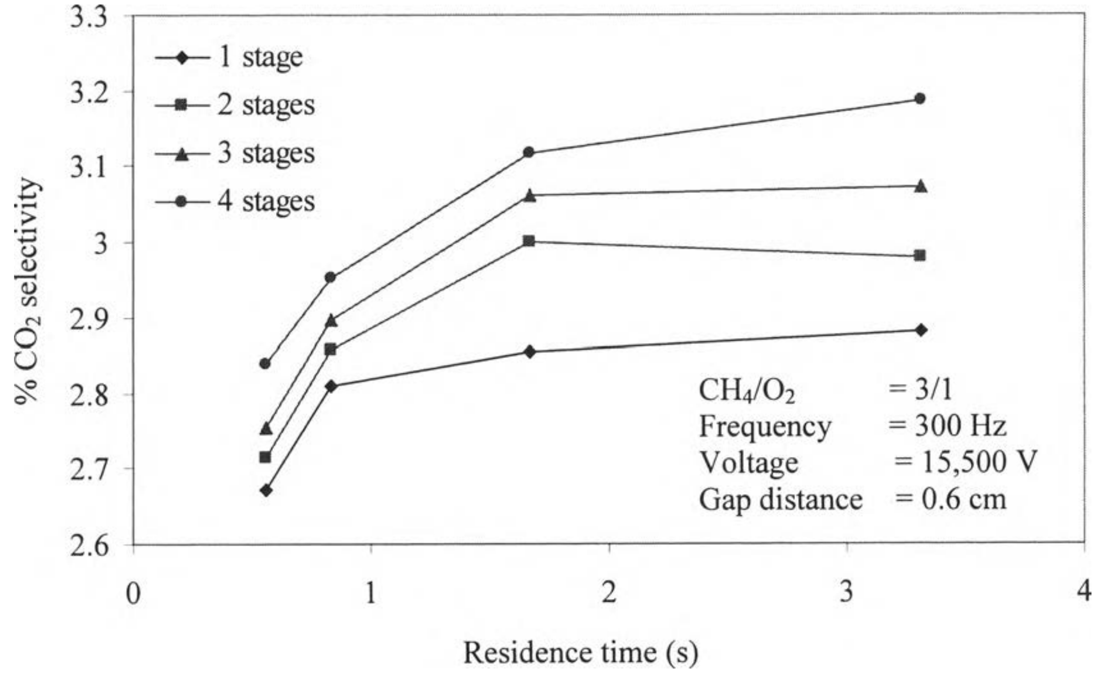

Figure 4.23 Effect of residence time on CO_2 selectivity at different stage numbers of the plasma system.

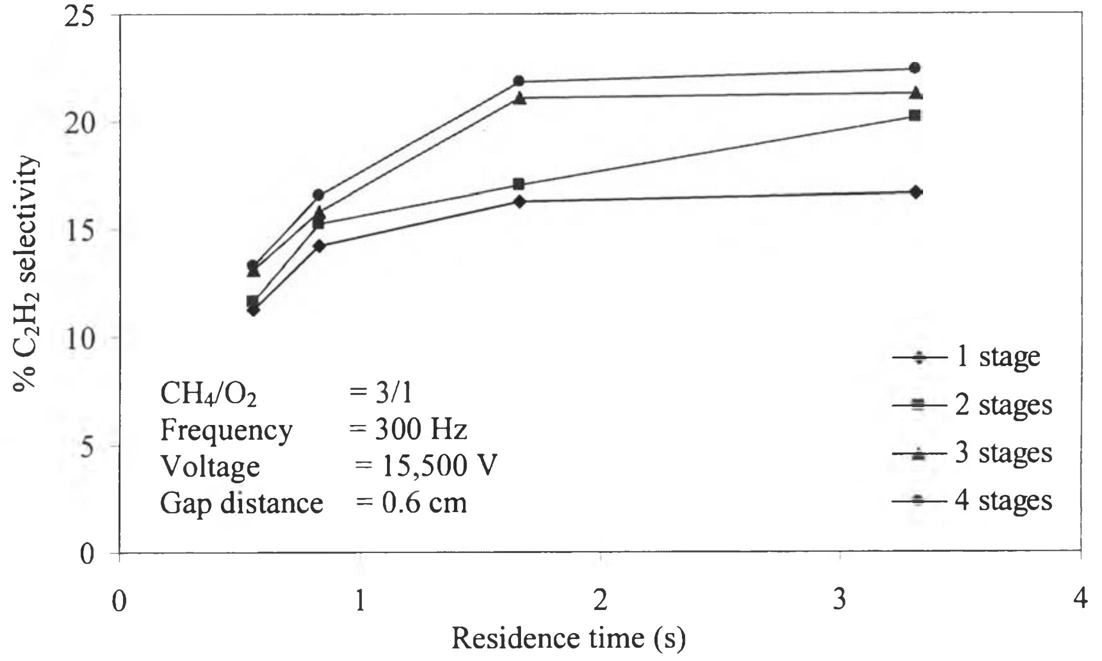

Figure 4.24 Effect of residence time on C_2H_2 selectivity at different stage numbers of the plasma system.

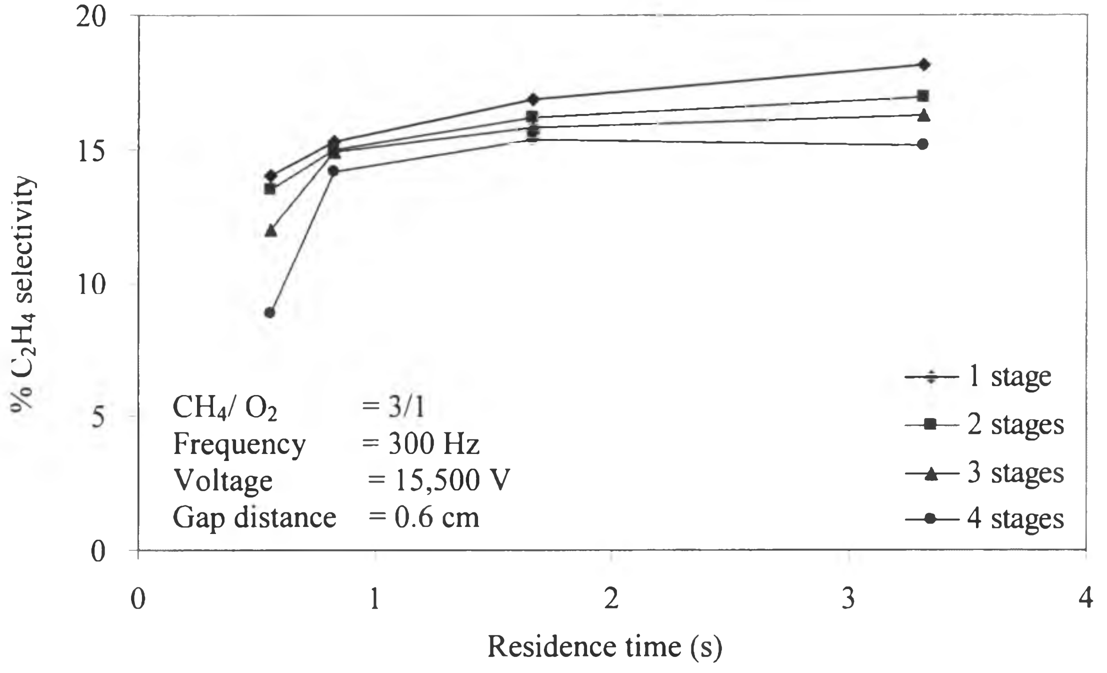

Figure 4.25 Effect of residence time on C_2H_4 selectivity at different stage numbers of the plasma system.

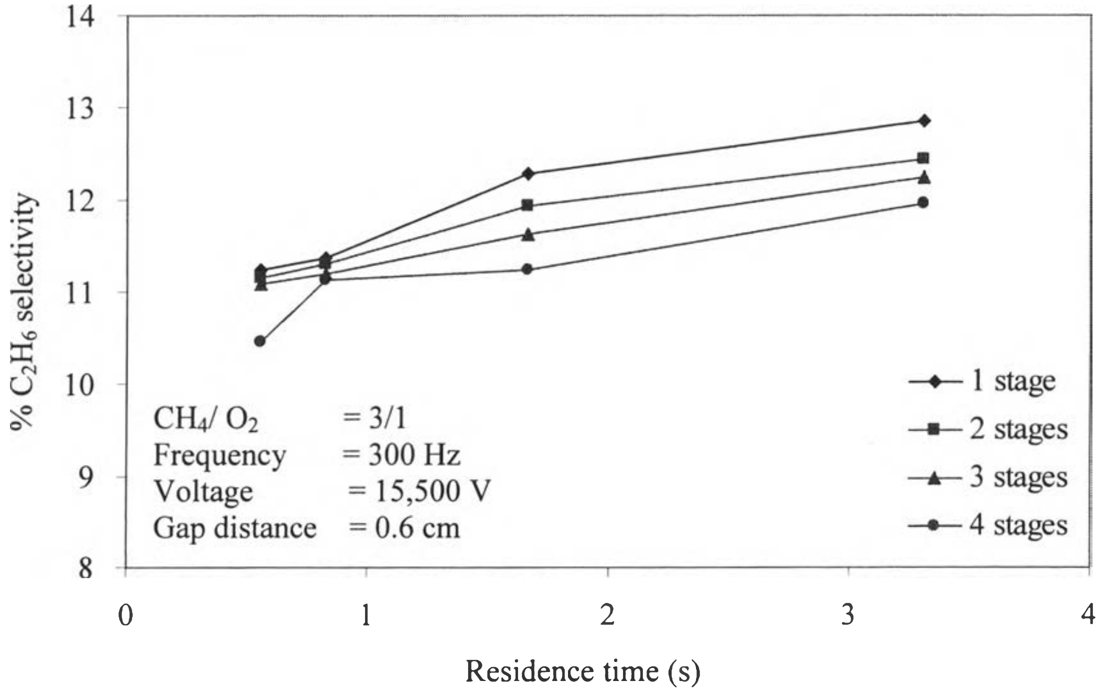

Figure 4.26 Effect of residence time on C_2H_6selectivity at different stage numbers of the plasma system.

4.4 Effect of Frequency

4.4.1 Effect on Methane and Oxygen Conversions

Figures 4.27 and 4.28 show the effects of frequency on CH_4 and O_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 of carbon deposit on the electrode surfaces. The conversions of CH_4 and O_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 CH_4 and O_2 , highest current is at this frequency as shown in Figure 4.29. At the highest current, there was the largest numbers of electrons generated in the system to initiate the reactions resulting in both maximum CH_4 and O_2 conversions. For any given frequency, both CH_4 and O_2 conversions increased with increasing stage numbers since the residence time increases.

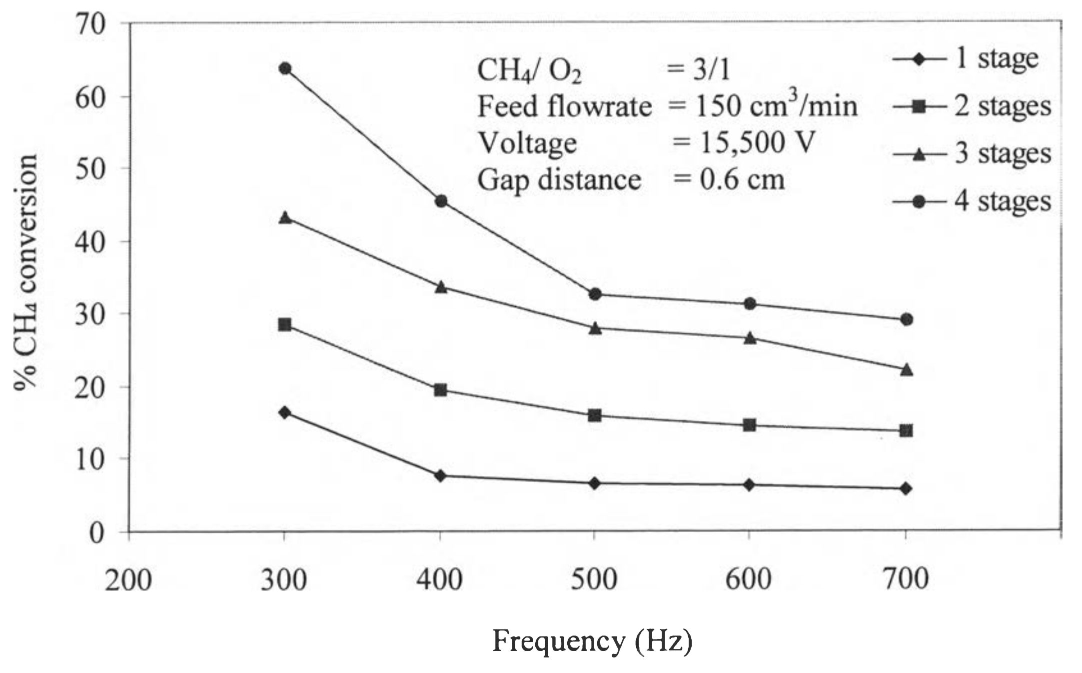

Figure 4.27 Effect of frequency on CH_4 conversion at different stage numbers of the plasma system.

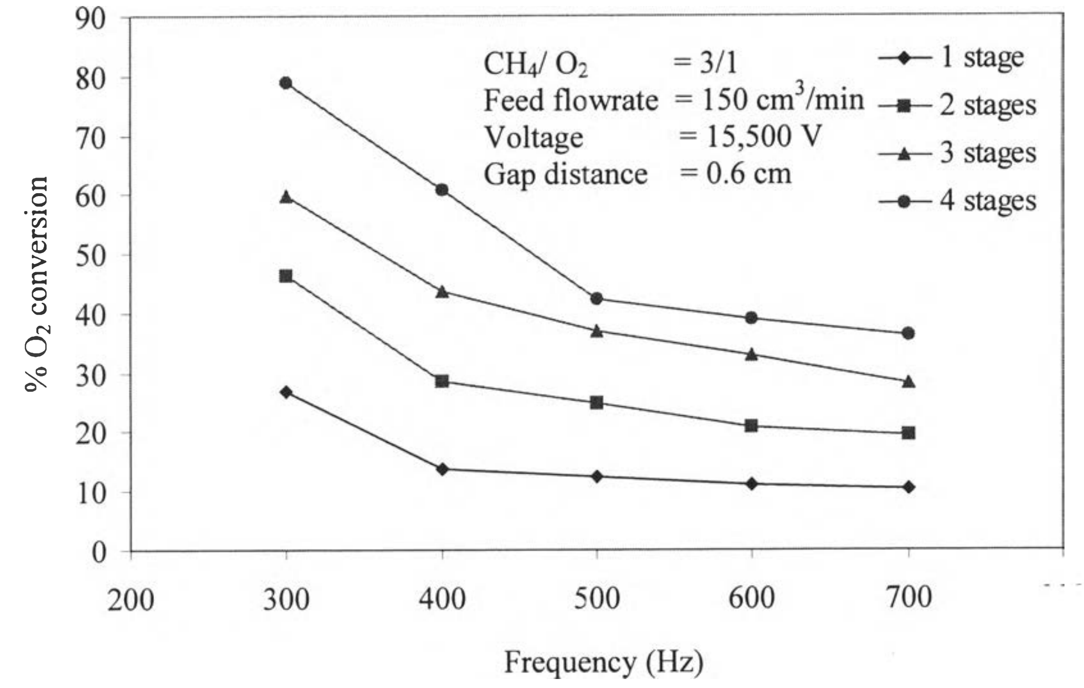

Figure 4.28 Effect of frequency on O_2 conversion at different stage numbers of the plasma system.

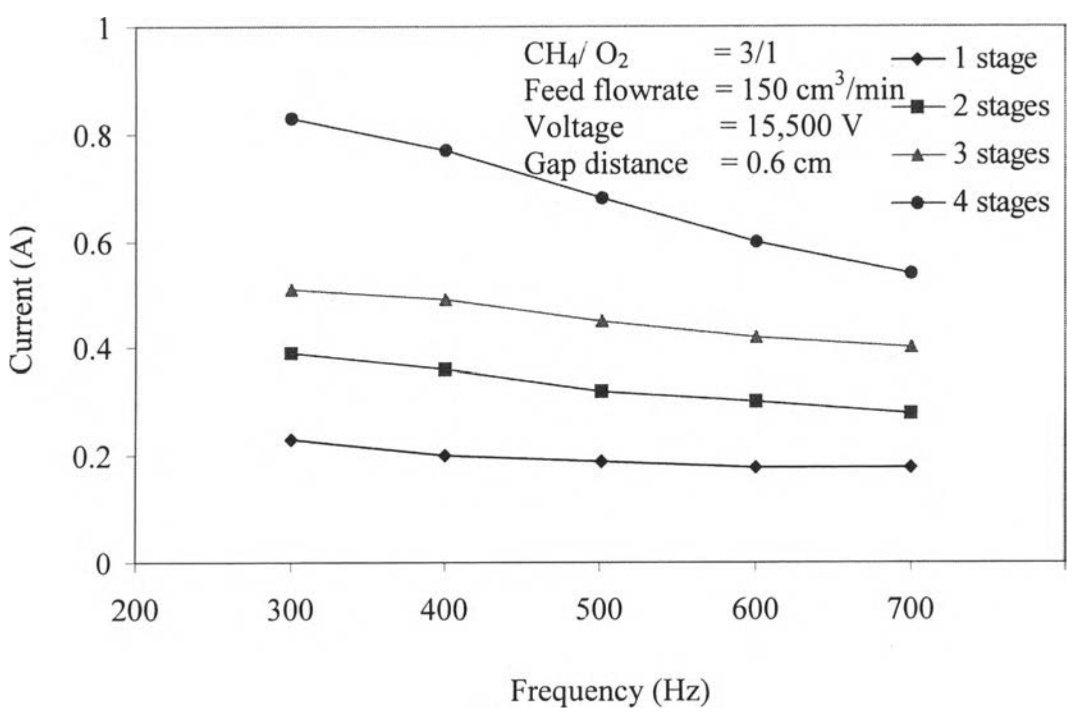

Figure 4.29 Effect of frequency on current at different stage numbers of the plasma system.

4.4.2 Effect on Product Selectivities

The effects of frequency on product selectivities are shown in Figures 4.30 to 4.35. The selectivities of CO and H_2 did not change significantly with increasing frequency but the selectivities of C_2H_2, C_2H_4 and C_2H_6 increased. 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 results reveal that the effect of frequency on the partial oxidation reaction of methane to produce synthesis gas is much greater than the oxidative coupling reaction of methane and dehydrogenation reaction.

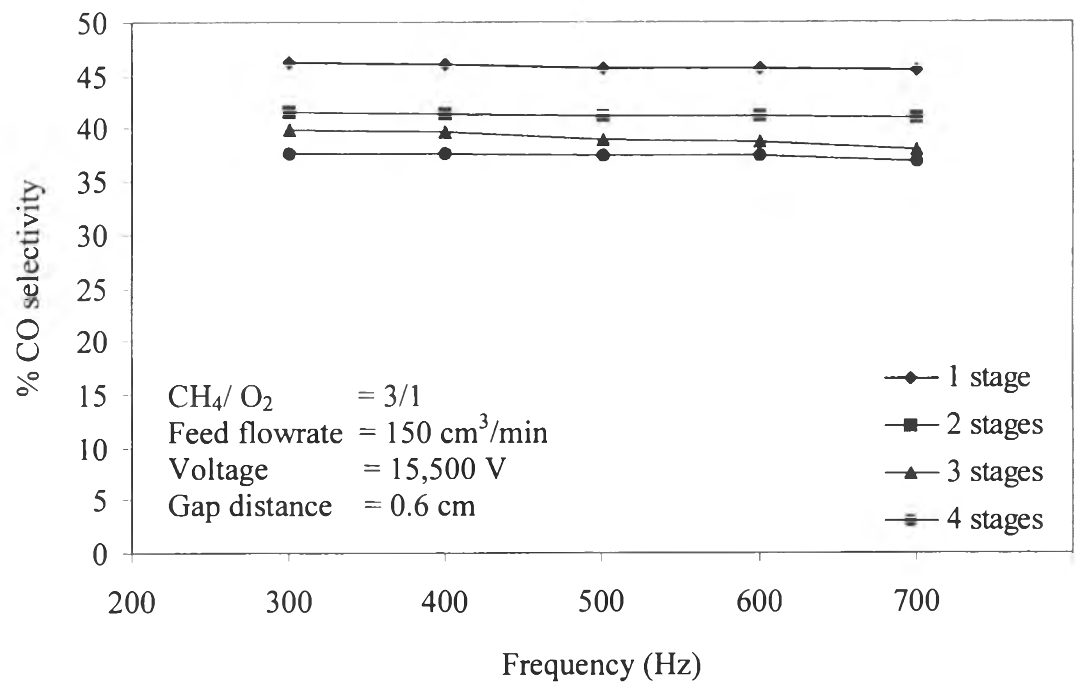

Figure 4.30 Effect of frequency on CO selectivity at different stage numbers of the plasma system.

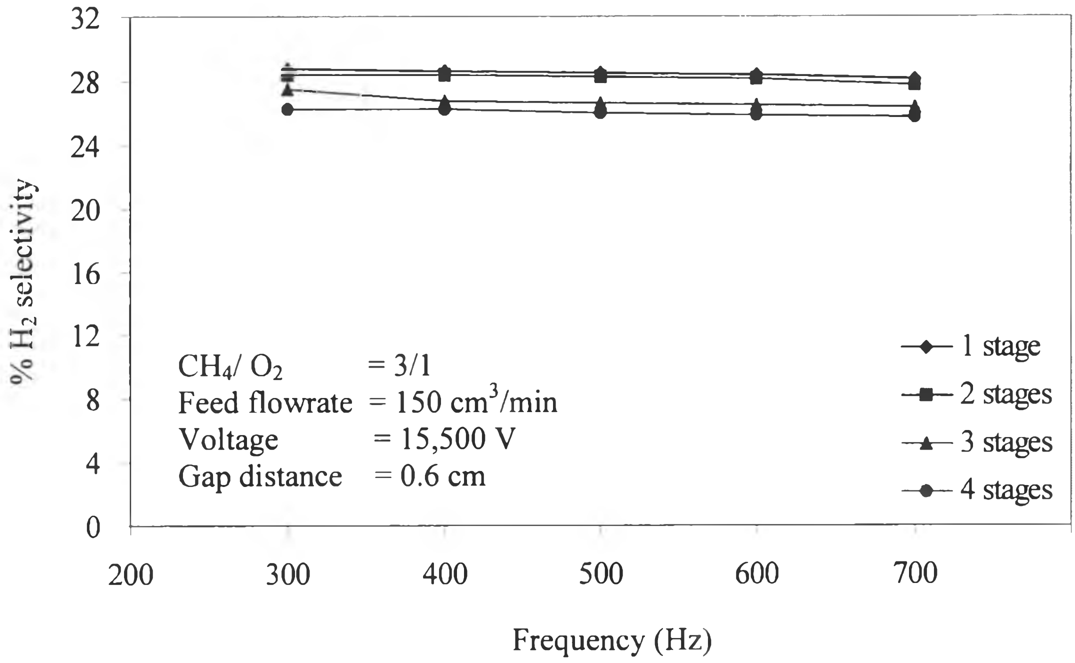

Figure 4.31 Effect of frequency on H_2 selectivity at different stage numbers of the plasma system.

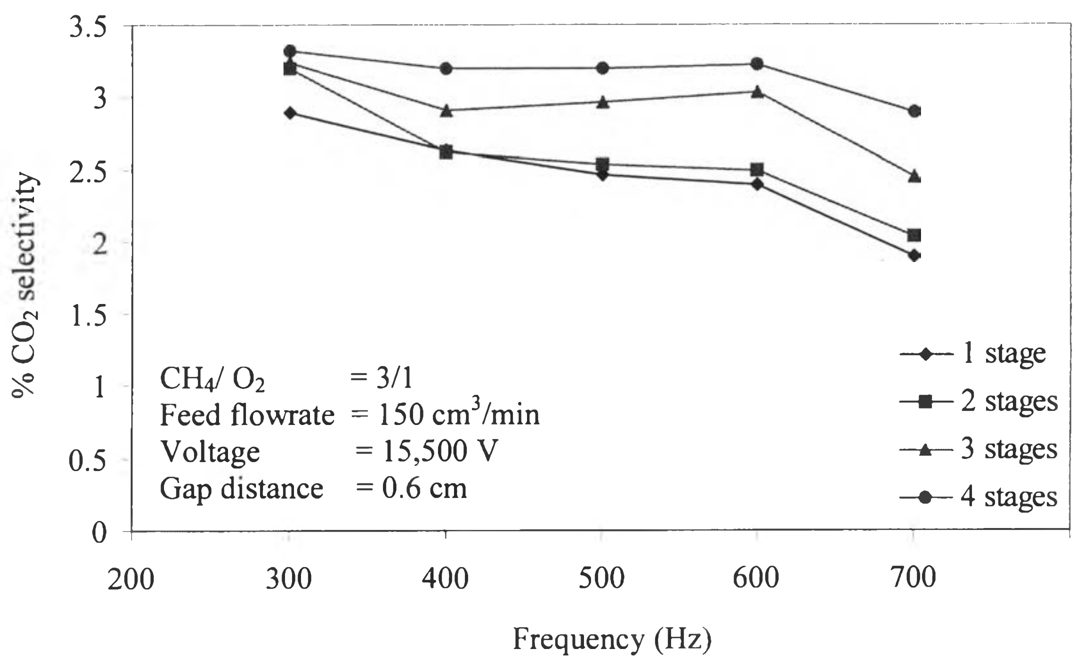

Figure 4.32 Effect of frequency on CO_2 selectivity at different stage numbers of the plasma system.

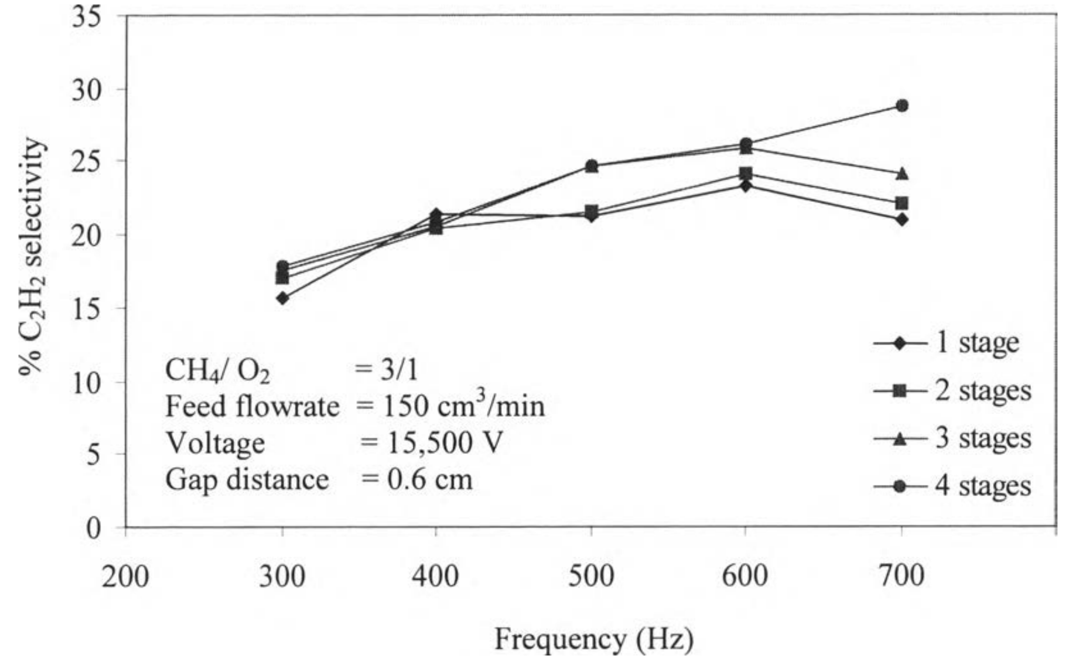

Figure 4.33 Effect of frequency on C_2H_2 selectivity at different stage numbers of the plasma system.

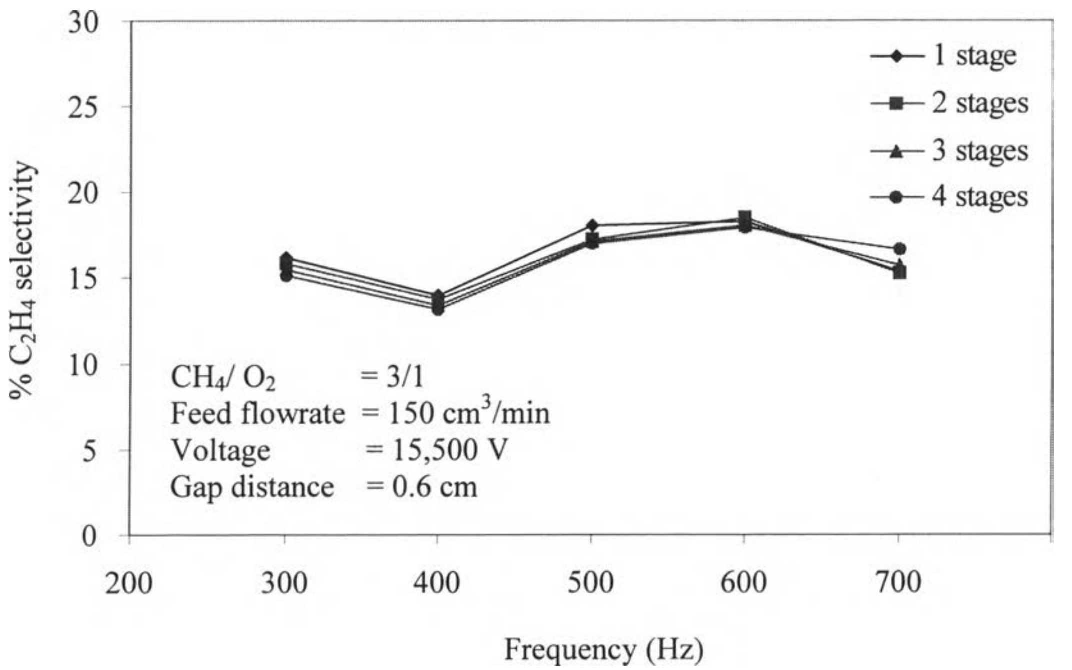

Figure 4.34 Effect of frequency on C_2H_4 selectivity at different stage numbers of the plasma system.

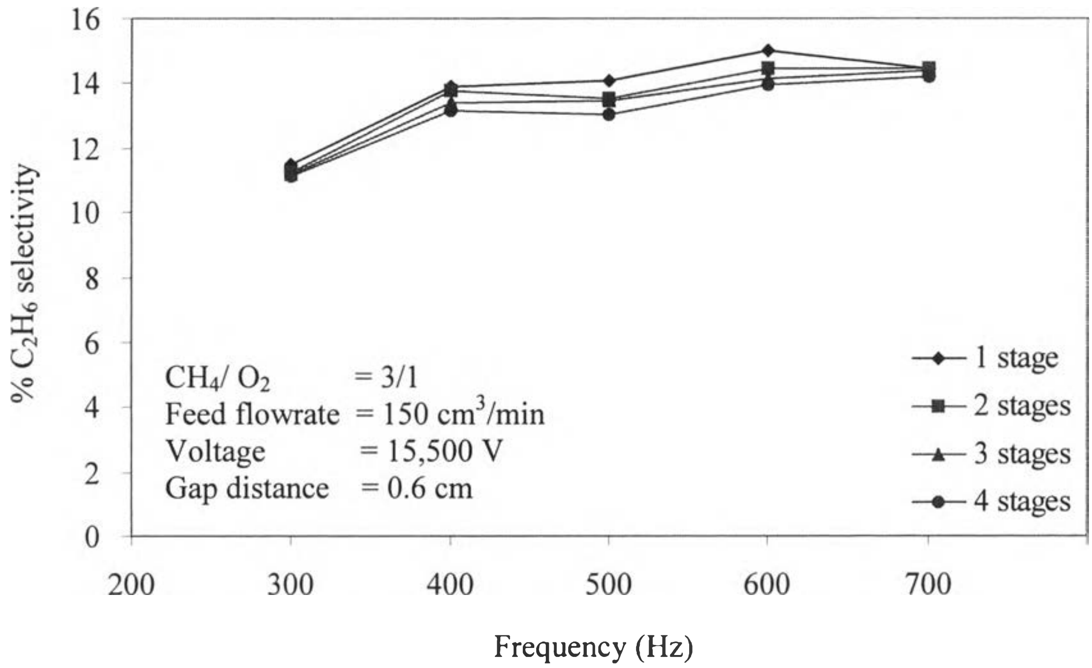

Figure 4.35 Effect of frequency on C_2H_6 selectivity at different stage numbers of the plasma system.

4.4.3 Effect on Power Consumption

The effect of frequency on power consumption is illustrated in Figure 4.36. The minimum power consumption was found at a frequency of 300 Hz. For a frequency below 300 Hz, the gliding arc system studied could not be operated because of carbon deposit on the surface of the electrodes. Since this carbon is electrically conductive, the current tends to flow almost entirely through these carbon deposits. This reduces the number of discharge streamers available to interact with the feed gas causing the reduction of both CH_4 and O_2 conversions. (Supat *et al.*, 2003)

In this study, 300 Hz is selected for next experiments because the highest CH_4 and O_2 conversions, and highest H_2 and CO selectivities as well as the lowest power consumption can be achieved.

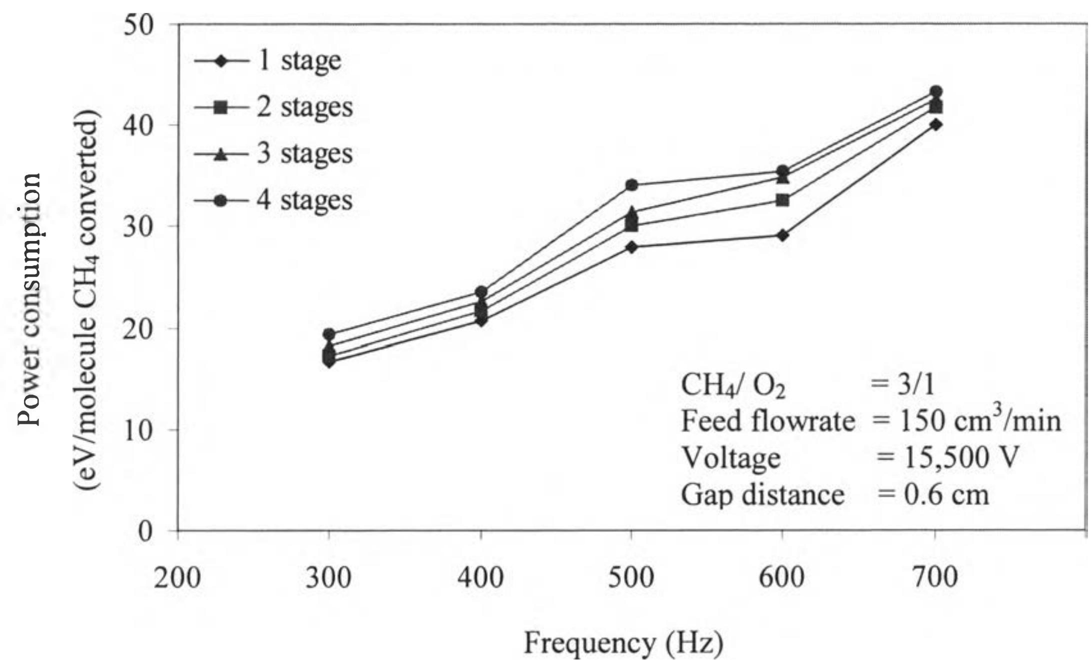

Figure 4.36 Effect of frequency on power consumption at different stage numbers of the plasma system.

4.5 Effect of Applied Voltage

4.5.1 Effect on Methane and Oxygen Conversions

The effects of applied voltage on CH_4 and O_2 conversions are shown in Figures 4.37 and 4.38, respectively. The applied voltage was varied from 10,000 to 19,000 V of the high side voltage since the break-down voltage of the study system is 10,000 V. For an applied voltage greater than 19,000 V, the system could not be operated because of the carbon deposit on the electrodes surfaces. In the contrast with increasing frequency, the CH_4 and O_2 conversions increased substantially with increasing applied voltage. The explanation is that a higher voltage results in higher current as shown in Figure 4.39. A higher current will give more available electrons to initiate the reactions because of the more opportunity of collision between CH_4 and O_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 CH_4 and O_2 conversions increased with increasing the stage number, as a result of increasing residence time. When the residence time increases the collision of CH_4 and O_2 molecules with the sufficient energy electrons increase.

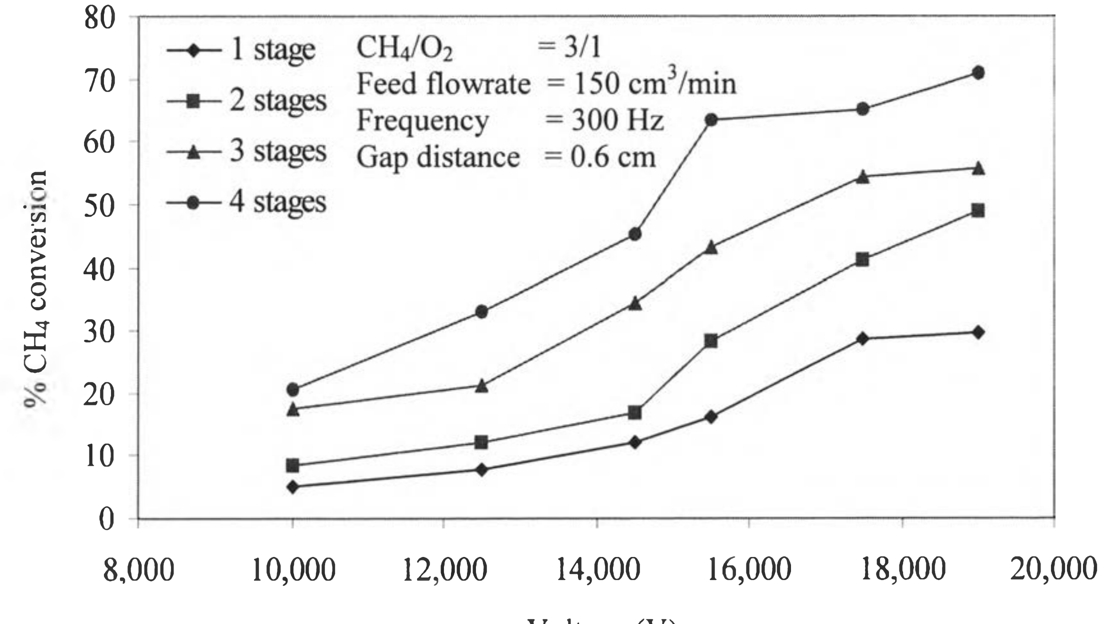

Figure 4.37 Effect of applied voltage on CH_4 conversion at different stage numbers of the plasma system.

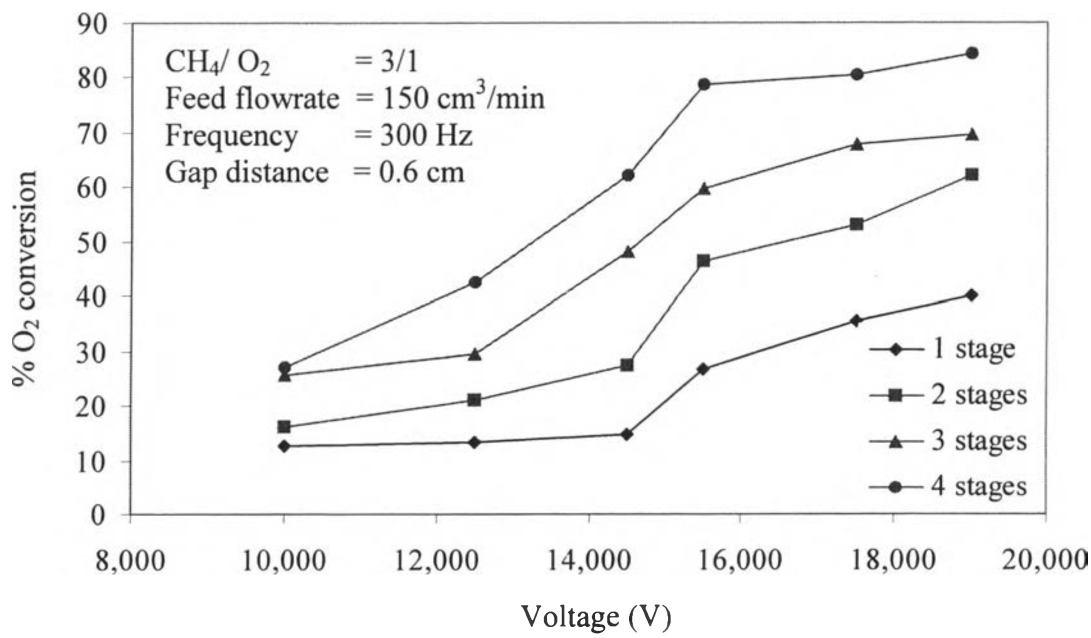

Figure 4.38 Effect of applied voltage on O_2 conversion at different stage numbers of the plasma system.

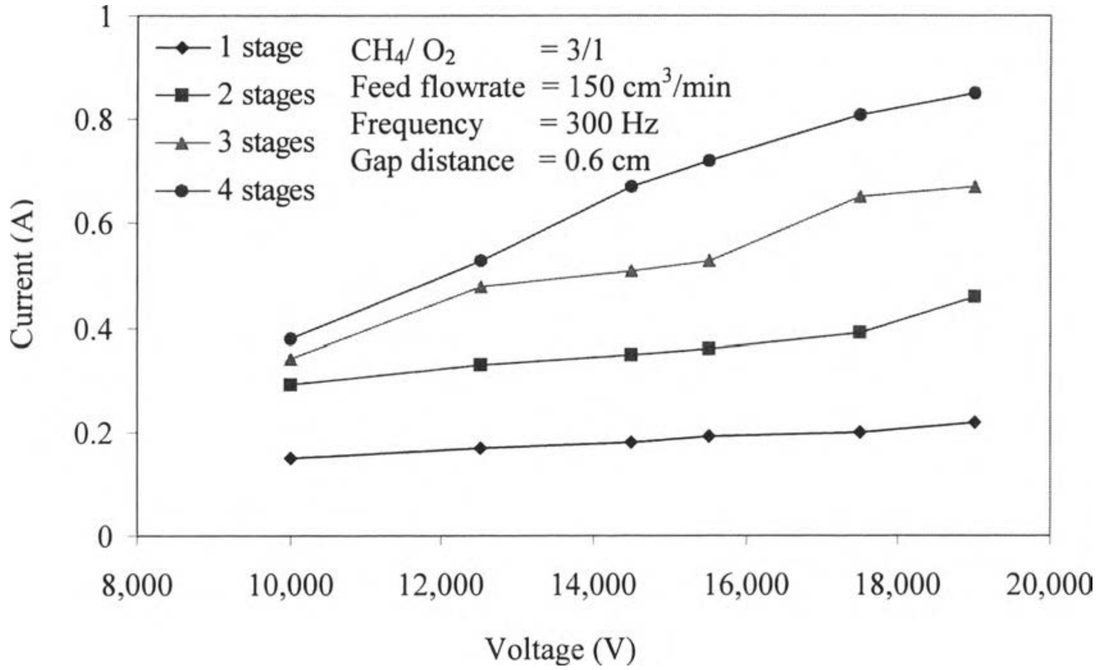

Figure 4.39 Effect of applied voltage on current at different stage numbers of the plasma system.

4.5.2 Effect on Product Selectivities

The effects of applied voltage on product selectivities are shown in Figures 4.40 to 4.45. The selectivities of CO, H_2, C_2H_4, and C_2H_6 decreased with increasing applied voltage and stage number, in contrast with the selectivities of CO_2 and C_2H_2. The results imply that CO, H_2, C_2H_4, and C_2H_6 are the primary products. When the applied voltage increases leading to higher average electron and O active species energy. Therefore the hydrocarbon primary products (C_2H_4, and C_2H_6) are further reacted known as dehydrogenation to form C_2H_2 and the CO oxidation is also increased. The oxidation reactions of CO and H_2 produce CO_2. An increase in the stage number corresponding to a higher residence time enhances both the CO oxidation and dehydrogenation reactions.

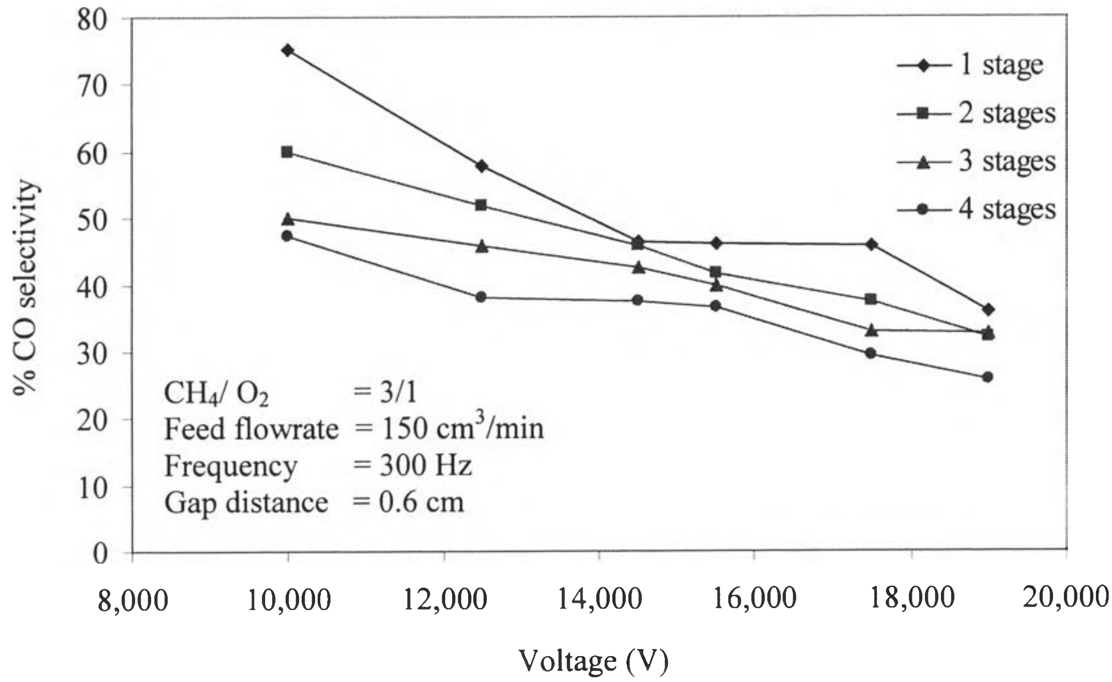

Figure 4.40 Effect of applied voltage on CO selectivity at different stage numbers of the plasma system.

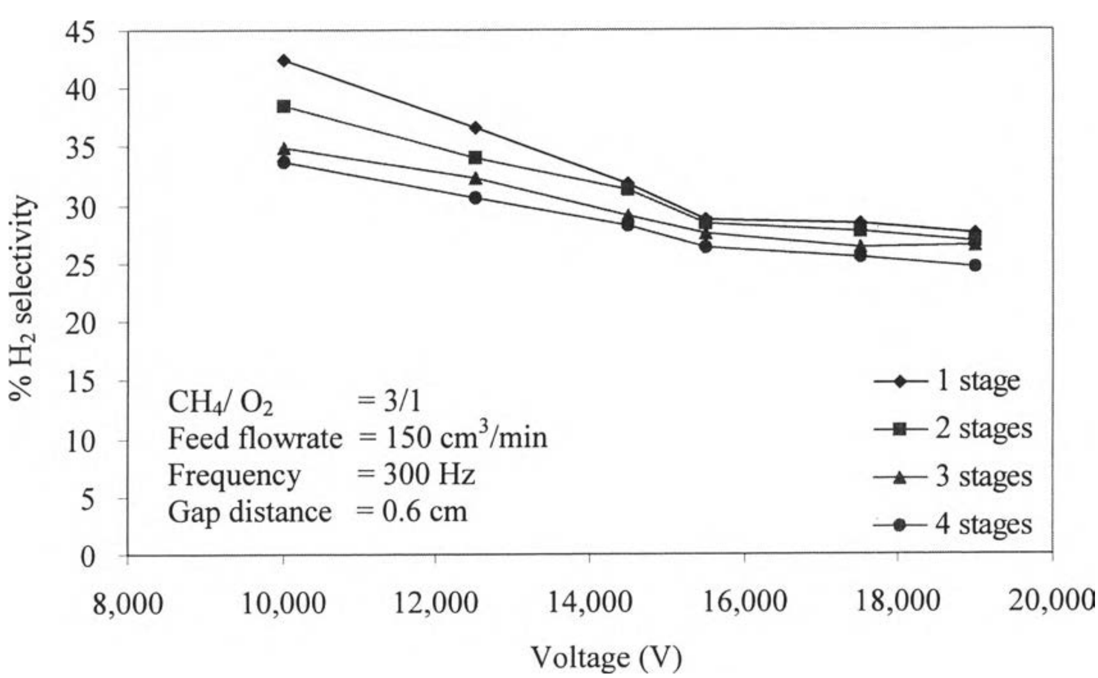

Figure 4.41 Effect of applied voltage on H_2 selectivity at different stage numbers of the plasma system.

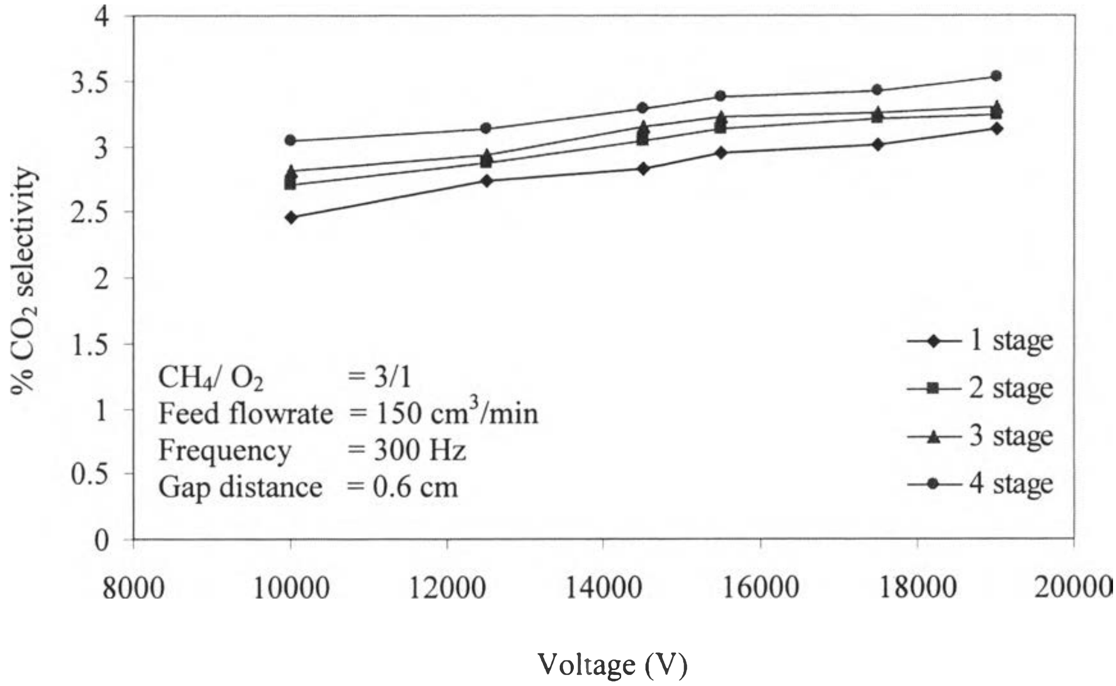

Figure 4.42 Effect of applied voltage on CO_2 selectivity at different stage numbers of the plasma system.

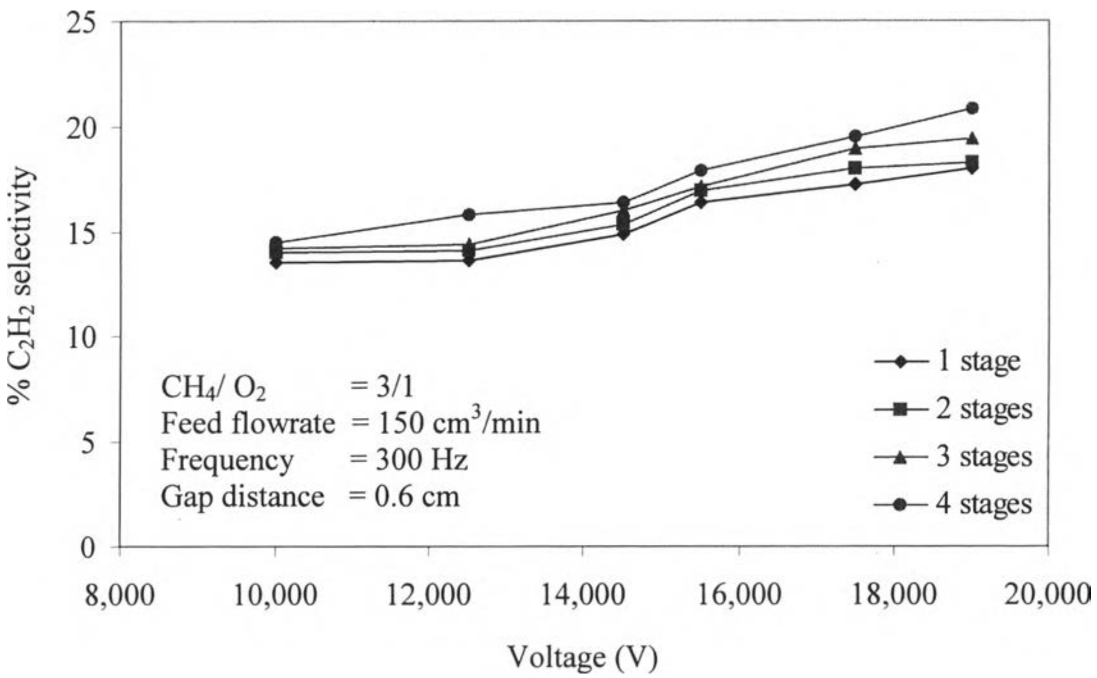

Figure 4.43 Effect of applied voltage on C_2H_2 selectivity at different stage numbers of the plasma system.

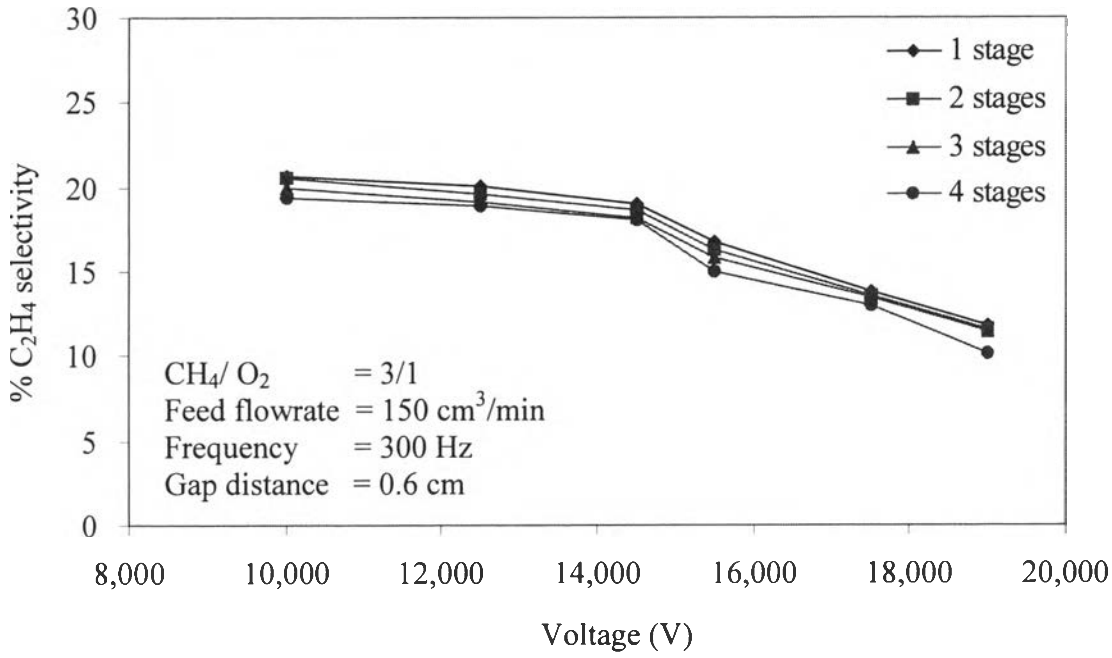

Figure 4.44 Effect of applied voltage on C_2H_4 selectivity at different stage numbers of the plasma system.

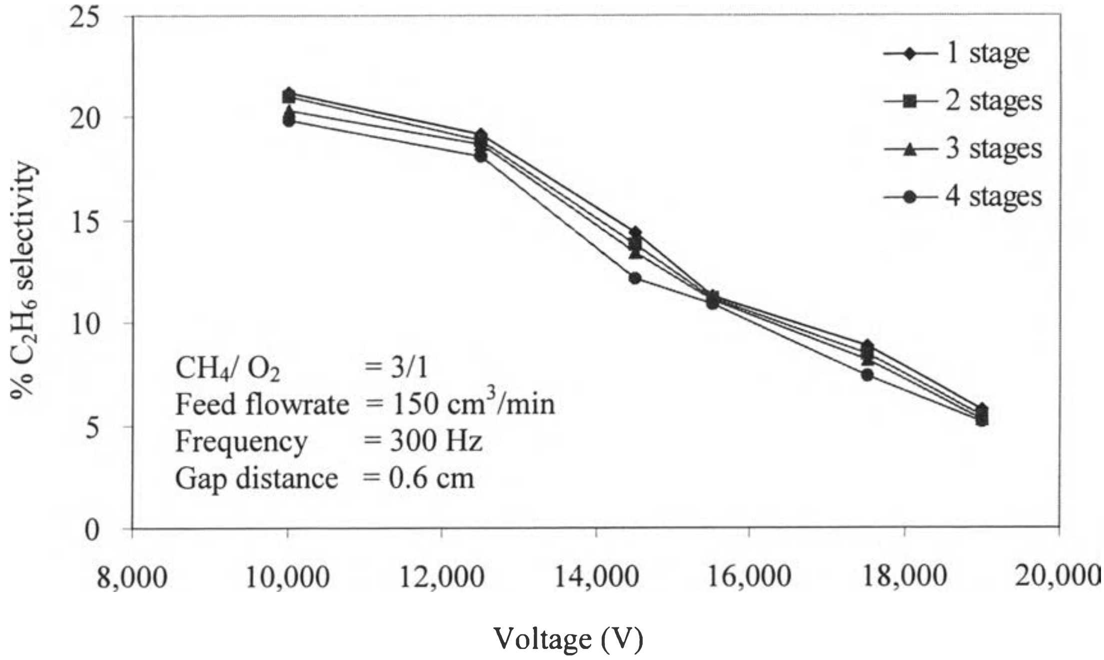

Figure 4.45 Effect of applied voltage on C_2H_6 selectivity at different stage numbers of the plasma system.

4.5.3 Effect on Power Consumption

Figure 4.46 shows the effect of applied voltage on the power consumption. With increasing applied voltage or stage number, the power consumption increased. At voltage higher than 19,000 V, the carbon deposit was found to form on the electrode surfaces. It is very interesting to point out that any optimum voltage depends upon what products are required. In this studied system, 14500 V was selected for next experiments since this voltage of 14,500 V gave high selectivities of the most products with a relatively low dehydrogenation reaction rate.

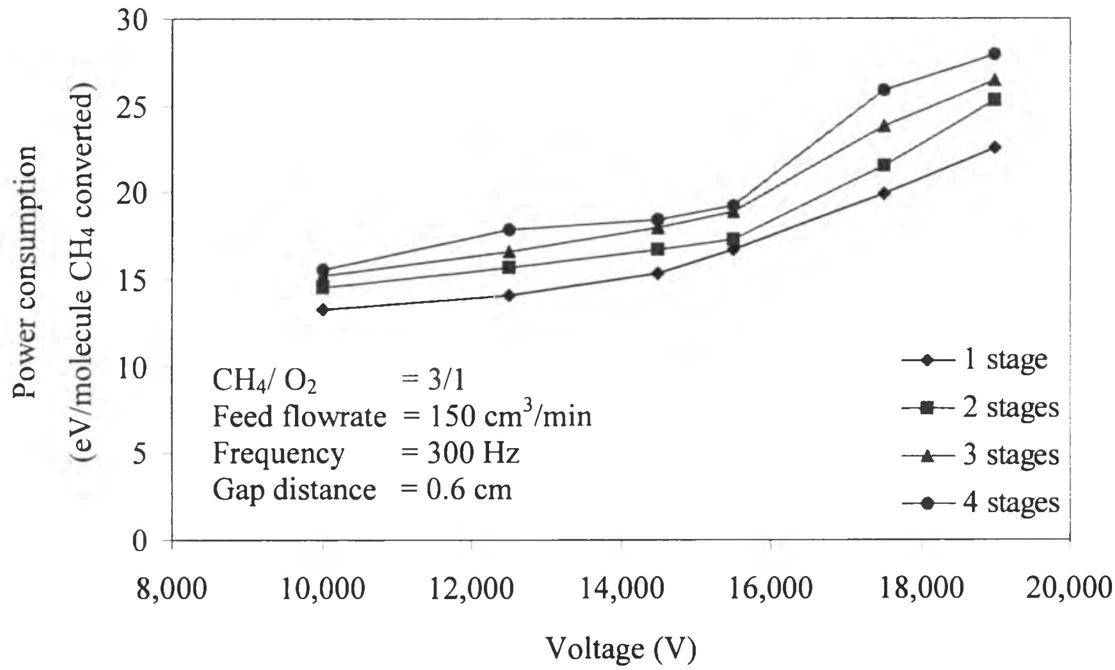

Figure 4.46 Effect of applied voltage on power consumption at different stage numbers of the plasma system.

4.6 Effect of Gap Distance

4.6.1 Effect on Methane and Oxygen Conversion

Figures 4.47 and 4.48 show the results of CH_4 and O_2 conversions, respectively. It is interesting note that a maximum gap distance of 8 mm could generate plasma under the studied system. The CH_4 and O_2 conversions increased with increasing the gap distance similar to that of the stage number. The increase in the gap distance or stage number simply increases the reaction volume or increasing residence time. As a result, these is more possibility for electrons to collide with CH_4 and O_2 molecules causing higher conversions of both reactants.

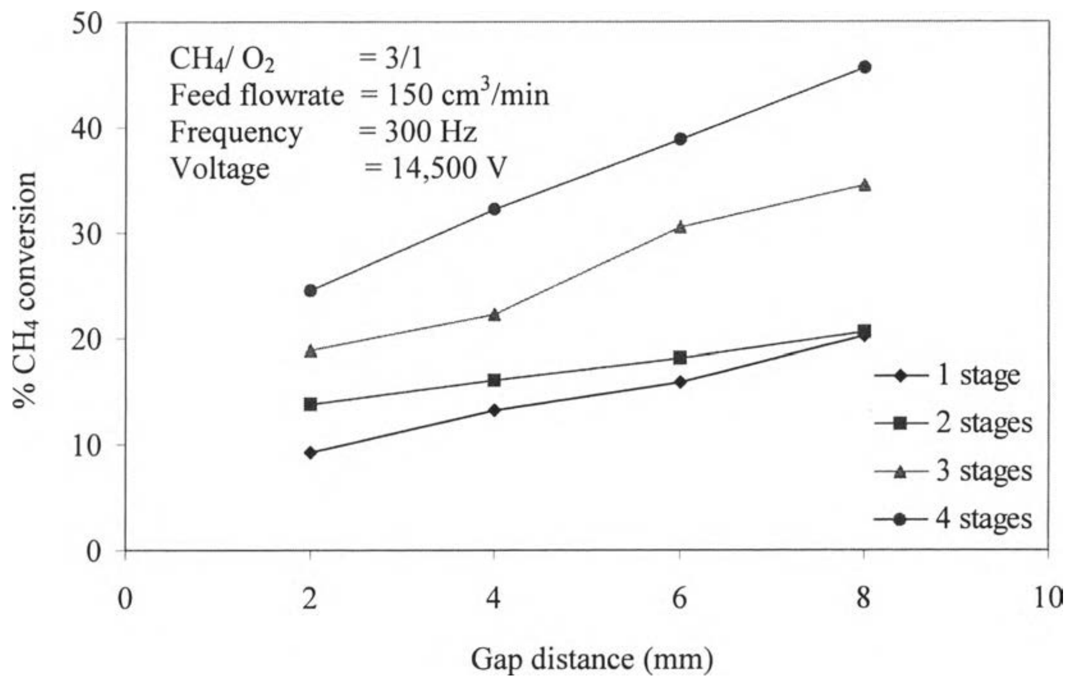

Figure 4.47 Effect of gap distance on CH_4 conversion at different stage numbers of the plasma system.

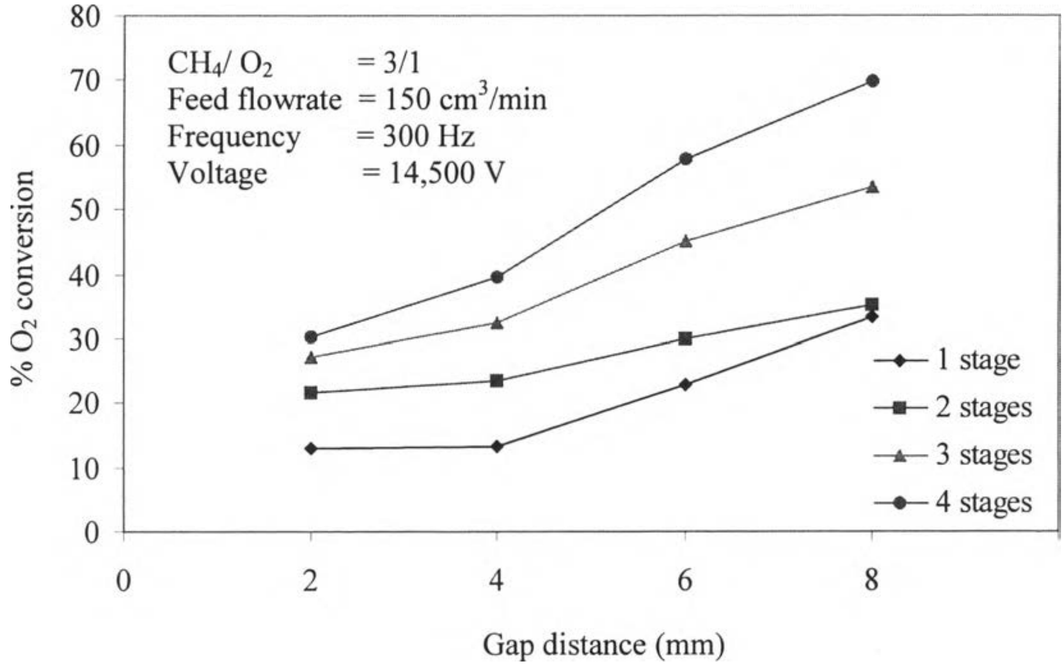

Figure 4.48 Effect of gap distance on O_2 conversion at different stage numbers of the plasma system.

4.6.2 Effect on Product Selectivities

The effects of gap distance on product selectivities are shown in Figures 4.49 to 4.54. With increasing the gap distance, the selectivities of CO, H_2, C_2H_4 and C_2H_6 decreased while the opposite trend was observed for the selectivities of CO_2 and C_2H_2. The explanation is that CO, H_2, C_2H_4 and C_2H_6 are the primary products. When either the gap distance or the stage number increases, the retention time simply increases leading to higher probability of electrons reacting with the primary products and O_2 molecules.

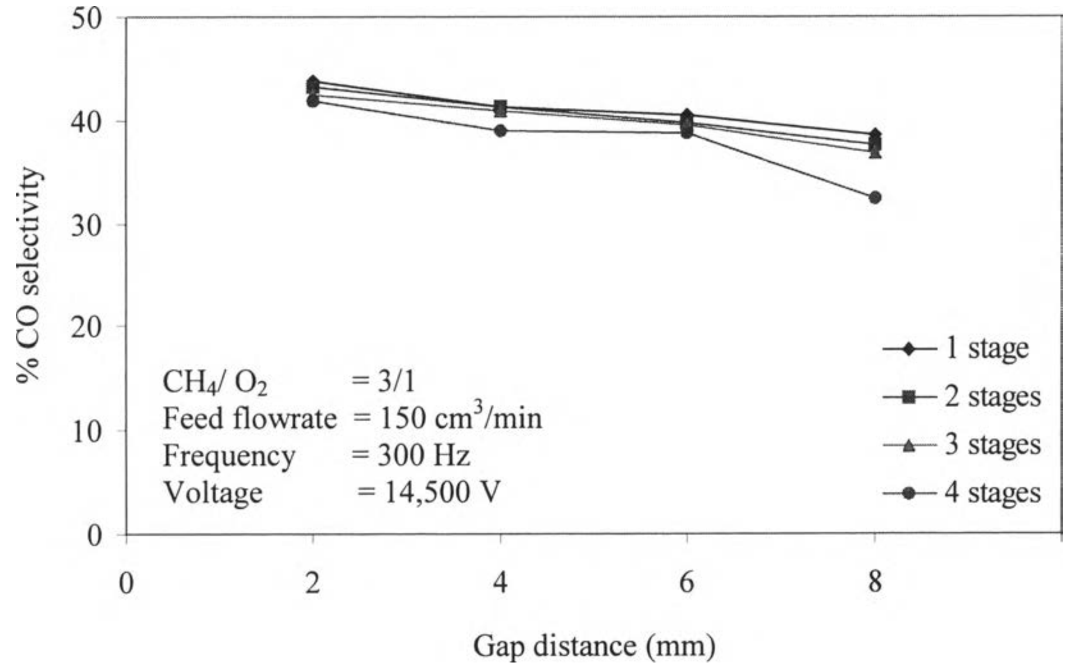

Figure 4.49 Effect of gap distance on CO selectivity at different stage numbers of the plasma system.

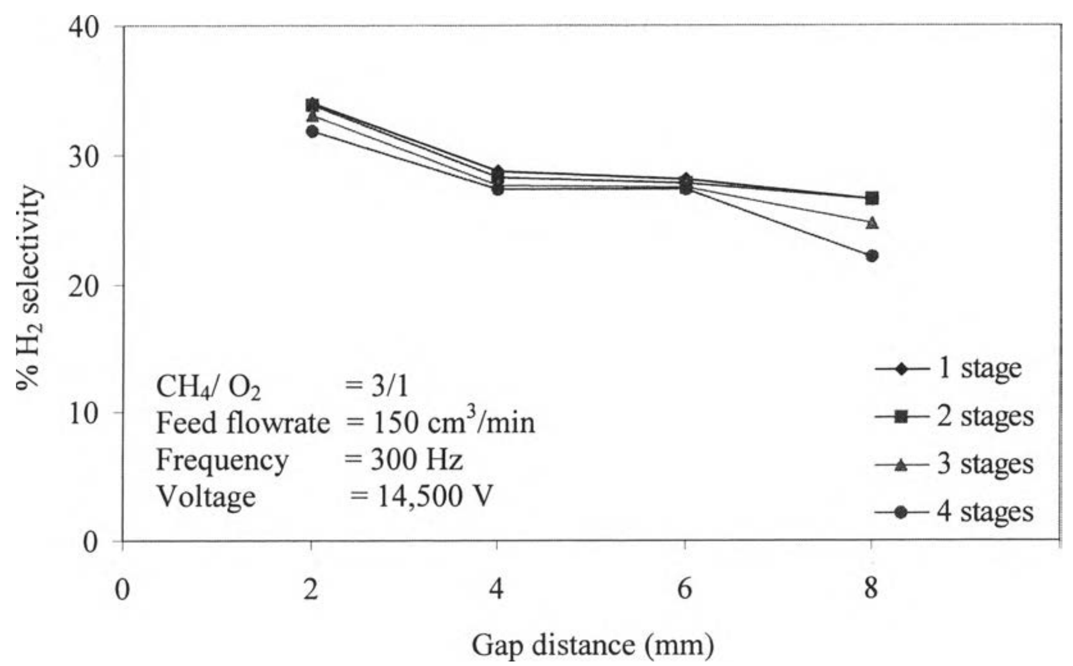

Figure 4.50 Effect of gap distance on H_2 selectivity at different stage numbers of the plasma system.

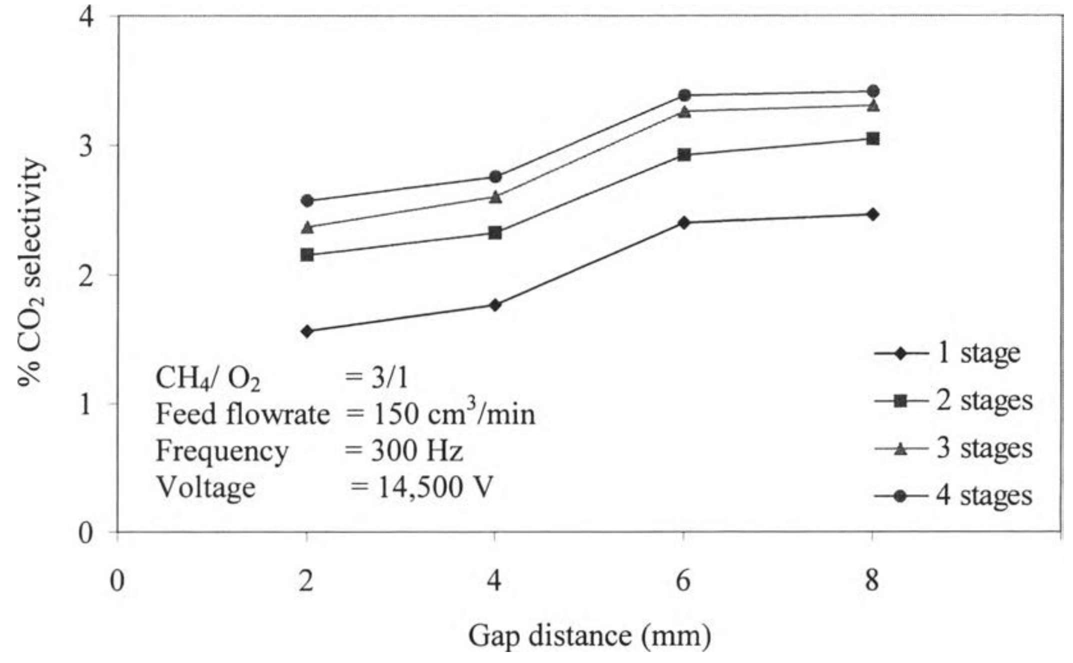

Figure 4.51 Effect of gap distance on CO_2 selectivity at different stage numbers of the plasma system.

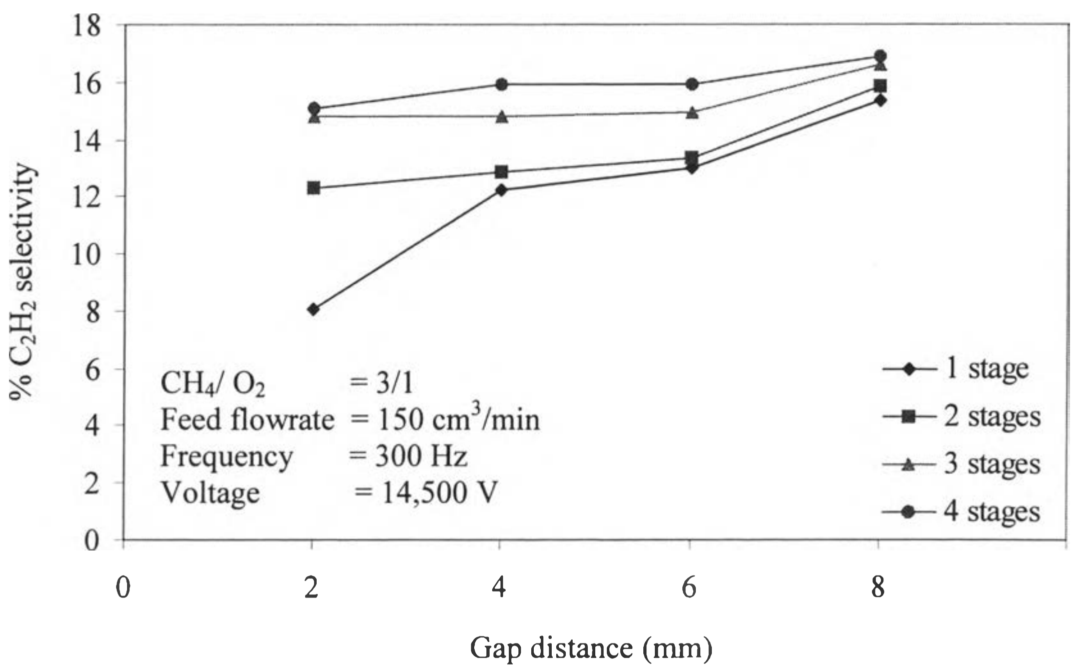

Figure 4.52 Effect of gap distance on C_2H_2 selectivity at different stage numbers of the plasma system.

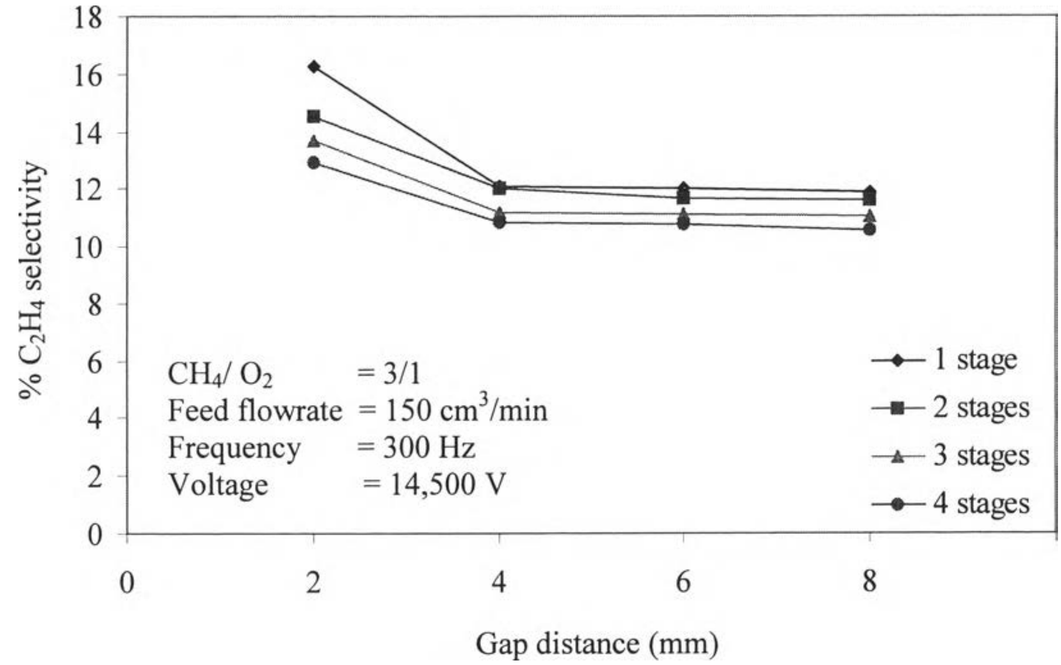

Figure 4.53 Effect of gap distance on C_2H_4 selectivity at different stage numbers of the plasma system.

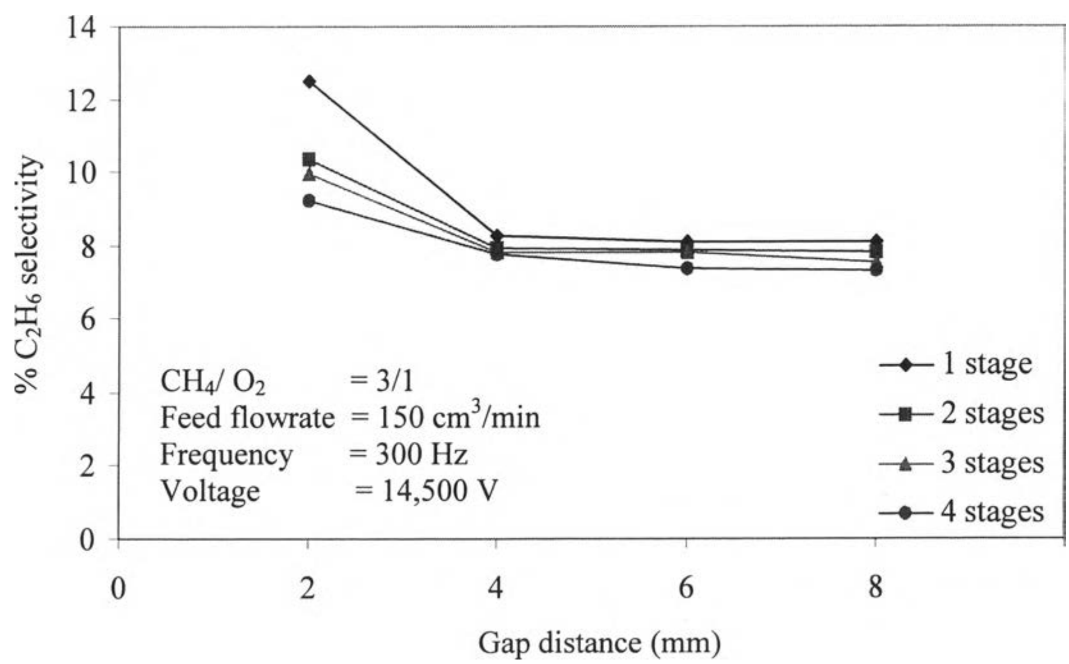

Figure 4.54 Effect of gap distance on C_2H_6 selectivity at different stage numbers of the plasma system.

4.6.3 Effect on Power Consumption

The power consumption increases with increasing gap distance and stage number as shown in Figure 4.55. It indicates that the energy distribution of electrons is improved in a higher gap distance as a result of increasing residence time. Figure 4.56 show the effect of stage number on the power consumption at different gap distance. At a gap distance of 0.6 cm from the stage one to the stage four the methane conversion increased from 15.89% to 38.90% but the power consumption decreased by 58.55%. The explanation for a significant decrease in the power consumption to convert a CH_4 molecule with increasing stage number is that a number of methane and oxygen molecule available becomes less and less with respect to a stage number of reactor. Another reason is that the products produced from the past reactor are further initiated by generated electrons leading to a higher power consumption with increasing stage number.

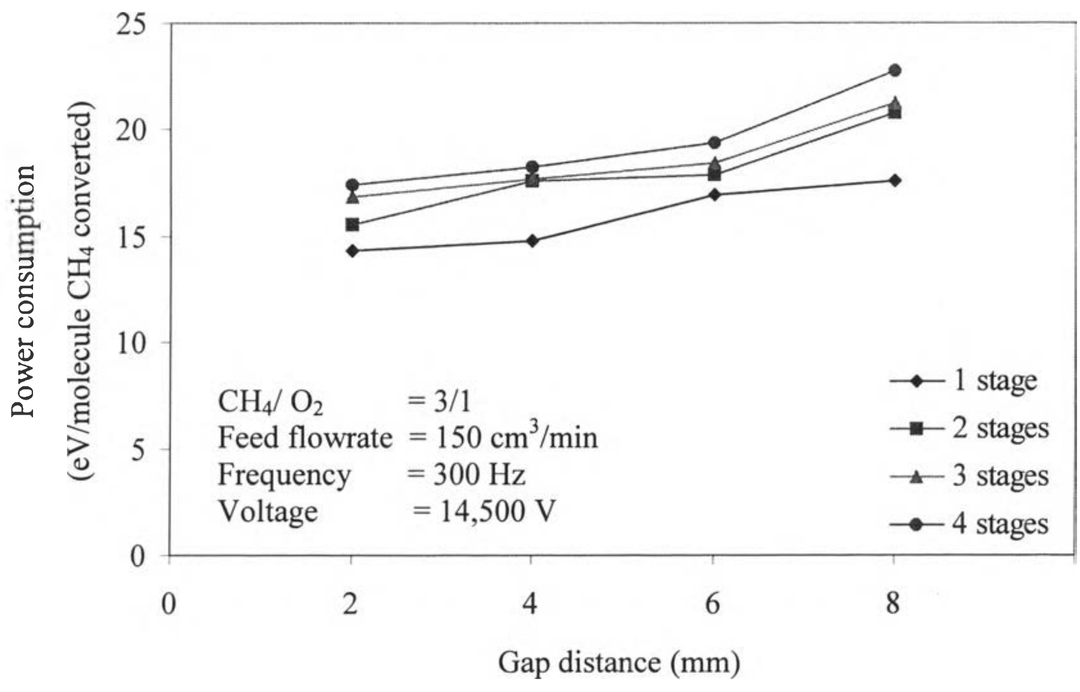

Figure 4.55 Effect of gap distance on power consumption at different stage numbers of the plasma system.

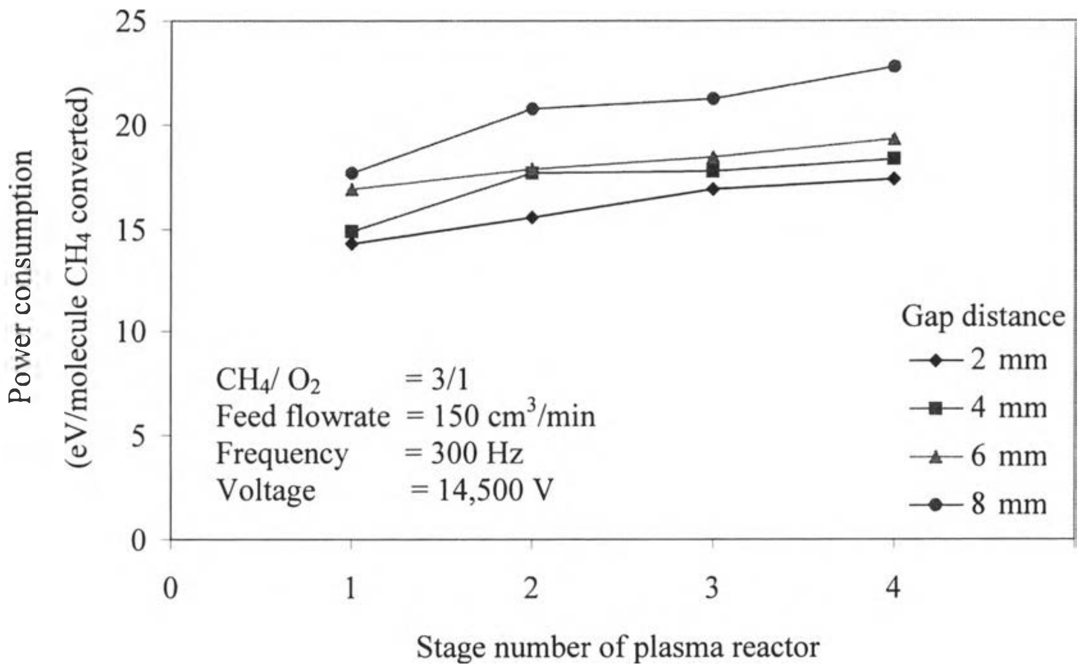

Figure 4.56 Effect of stage number on power consumption at different gap distance.