# CHAPTER IV
RESULTS AND DISCUSSION

4.1 Catalyst Characterization

The BET surface area of TiO_2 coated on glass wool was 27.4 m^2/g compared to 50 and 4.2 m^2/g for Degussa P-25 and glass wool, respectively. As shown in Figure 4.1, the crystalline phase of TiO_2 after annealing at 300°C and treating with He-plasma for 2 hr was the same as the original TiO_2, Degussa P-25. The TiO_2 particles agglomerated as layers and big clusters on the glass fibers, Figure 4.2.
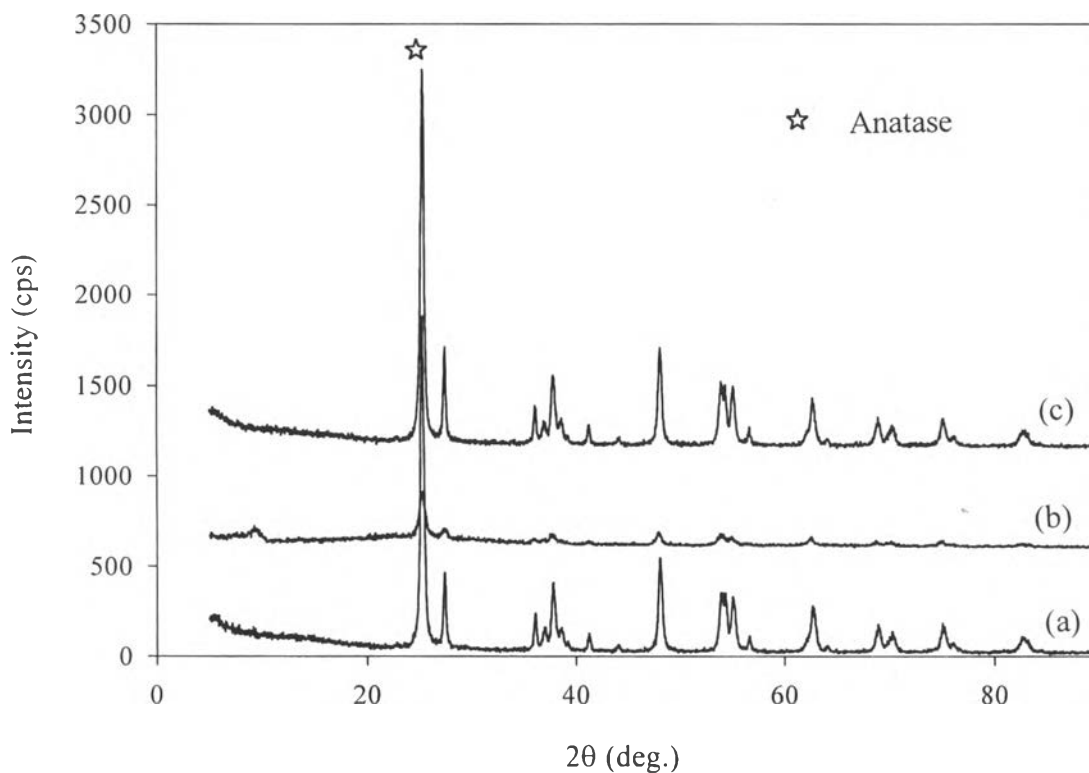
Figure 4.1 XRD patterns of (a) raw TiO_2, (b) TiO_2 coated on glass wool and (c) TiO_2 treated by He-plasma
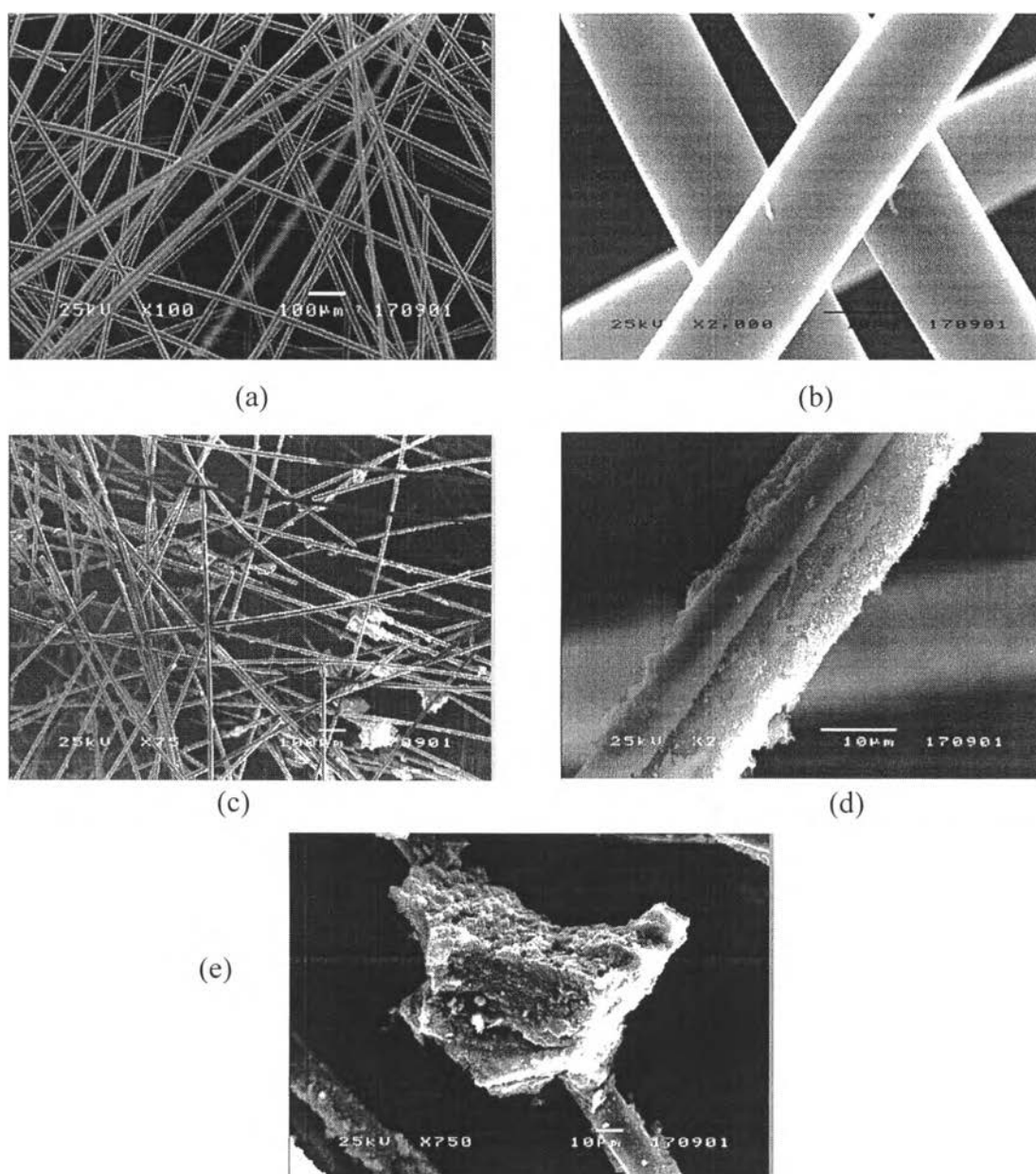
Figure 4.2 (a)-(b) Glass wool after dewaxing, (c)-(e) TiO_2 coated on glass wool

4.2 Effect of the Feed Flow Rate

4.2.1 Effect on the Ethylene and Oxygen Conversion

Figure 4.3 shows the effect of the feed flow rate on the ethylene and oxygen conversion. As an increase in the flow rate corresponds to a decrease in residence time, both ethylene and oxygen conversion decreases with the increase of the total the gas flow rate of the feed gas from 20 to 80 ml/min. The conversion of

ethylene is much higher than that of oxygen. This is probably due to the ozone decomposition to oxygen during analysis by GC.
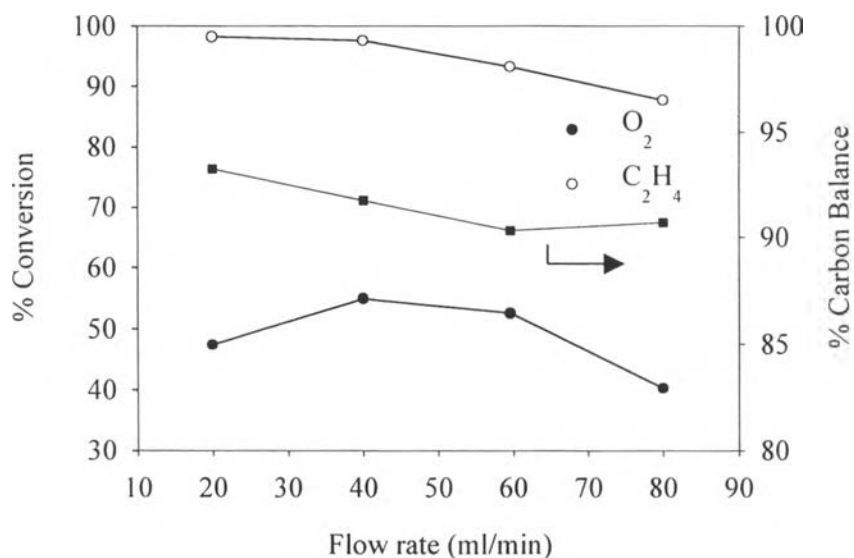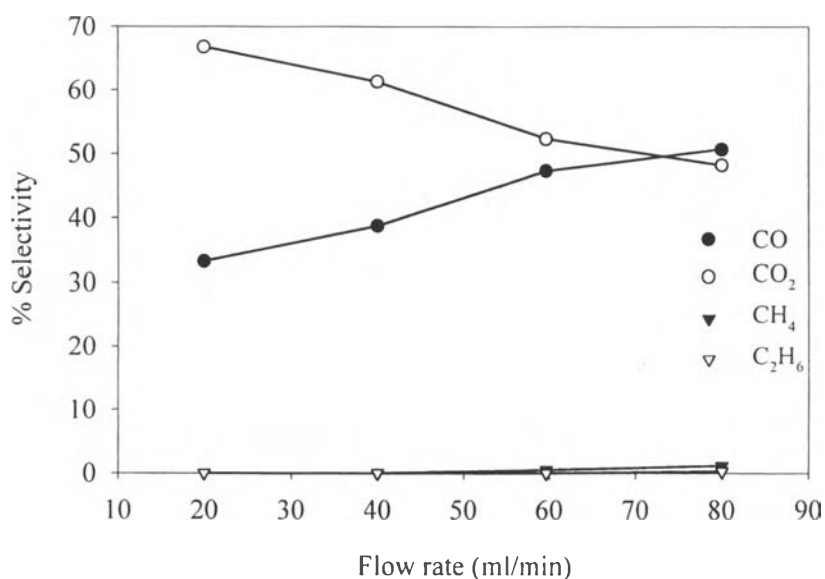

Figure 4.3 Effect of the feed flow rate on the ethylene and oxygen conversion at 3.5 W and 400 Hz

4.2.2 Effect on the Product Selectivity

The effect of the flow rate on the product selectivity is shown in Figure 4.4. The CO_2 selectivity decreases as the total flow rate increases whereas the CO selectivity increases. It can be observed that the CO_2 selectivity is higher and the CO selectivity is lower as the total flow rate decreases. It can be explained that the higher gas flow rate suppresses the possibility of collision between electrons and oxygen molecules, which then lowers the active species of oxygen; therefore, the oxidation of carbon monoxide is reduced. In addition, methane and ethane were detected when the flow rates were 60 and 80 ml/min. Since the C-H bond energy is higher than the C=C bond, the C-C cleavage in ethylene should be the initial step of the ethylene destruction. As proposed by Liao *et al.* (2002), the produced methyl radicals were more feasible to form methane by abstracting hydrogen and ethane by coupling with other methane radicals as the follows:

$$C_2H_4 \quad + \quad O^\bullet \quad \longrightarrow \quad CH_3^\bullet \quad + \quad CHO^\bullet \tag{4.1}$$

$$CH_3^\bullet \quad + \quad CHO^\bullet \quad \longrightarrow \quad CH_4 \quad + \quad CO \tag{4.2}$$

$$CH_3^\bullet \quad + \quad H^\bullet \quad + \quad M \quad \longrightarrow \quad CH_4 \quad + \quad M \tag{4.3}$$

$$2\,CH_3^\bullet \quad \longrightarrow \quad C_2H_6 \tag{4.4}$$

$$CHO^\bullet \quad + \quad e^- \quad \longrightarrow \quad H^\bullet \quad + \quad CO \quad + e^- \tag{4.5}$$

$$CO \quad + \quad OH^\bullet \quad \longrightarrow \quad CO_2 \quad + \quad H^\bullet \tag{4.6}$$

$$CO \quad + \quad O^\bullet \quad + \quad M \quad \longrightarrow \quad CO_2 \quad + \quad M \tag{4.7}$$

where M is a third collision partner : O_2, O_3 and O

**Figure 4.4** Effect of the feed flow rate on the product selectivity at 3.5 W and 400 Hz

4.3 Effect of the Frequency

4.3.1 Effect on the Ethylene and Oxygen Conversion

Figure 4.5 illustrates the effect of the frequency on the ethylene and oxygen conversions. The conversions of ethylene and oxygen are almost constant in the studied frequency range of 200 to 500 Hz. It can be explained that the current, which corresponds to the amount of electrons, is almost constant with increasing frequency as shown in Figure 4.6.
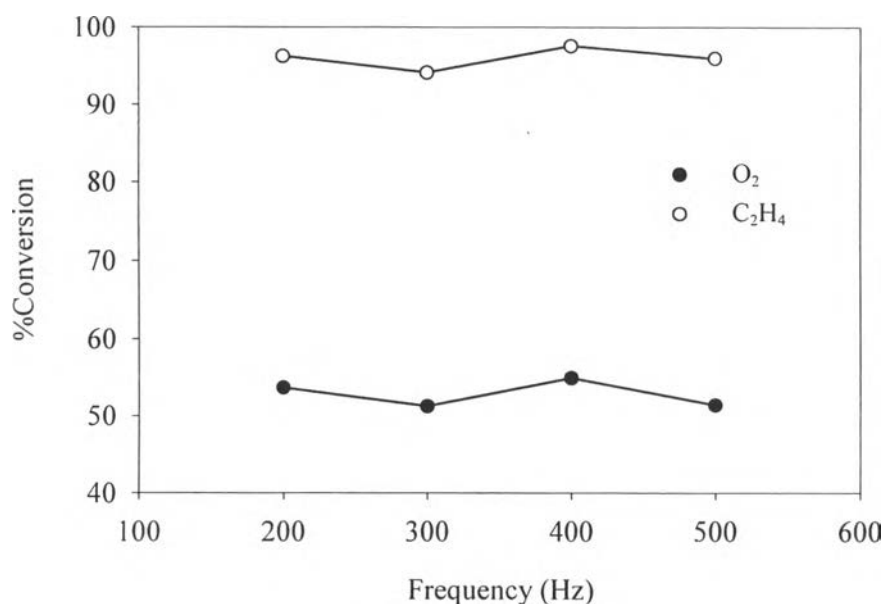
Figure 4.5 Effect of the frequency on the ethylene and oxygen conversion at 3.5 W and the total gas flow rate of 40 ml/min
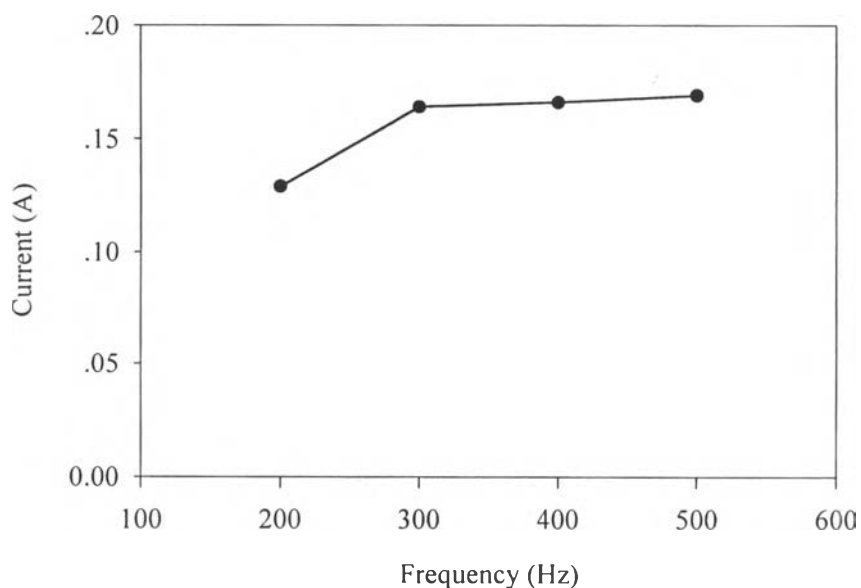

Figure 4.6 Effect of frequency on the current generated at 3.5 W

4.3.2 Effect on the Product Selectivity

The effect of the applied frequency on the product selectivity is shown in Figure 4.7. Similar to the effect of frequency on the conversion of O_2 and C_2H_4, the frequency has little effect on the product selectivity. However, the CO_2 selectivity is higher when the frequency is lower than 300 Hz. That is

because the energy of electrons at near the upper electrode is the highest and carbon layers are formed on the upper electrode. As a result, more electrons on the electrodes are consumed directly by the carbon layer and the oxidation of the deposited carbon is more complete.
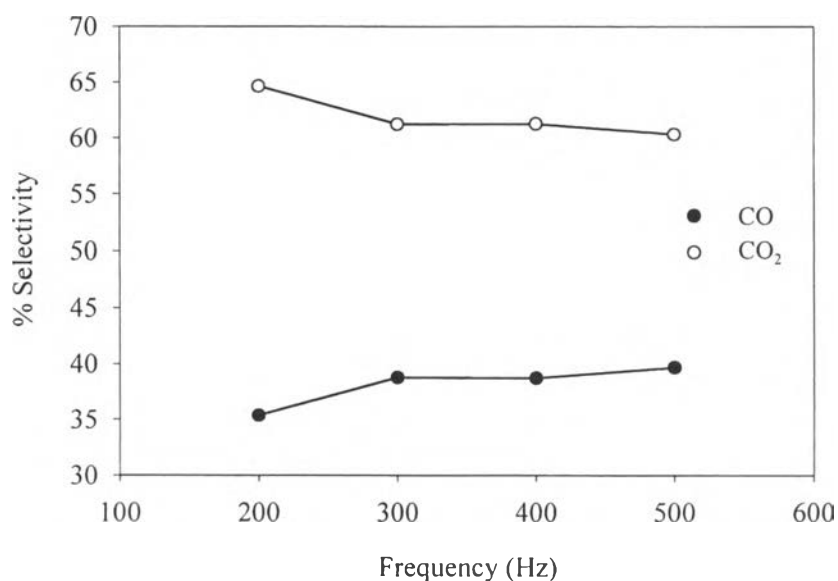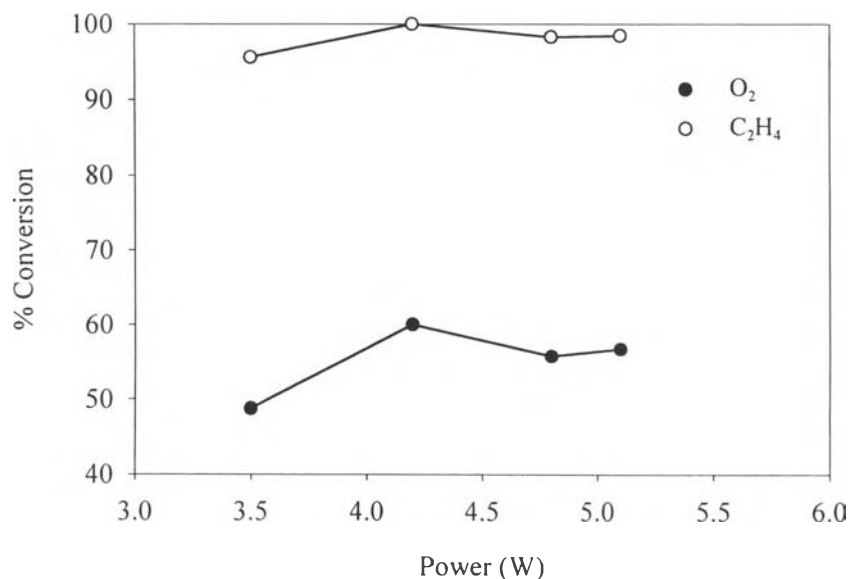

Figure 4.7 Effect of the frequency on the product selectivity at 3.5 W and the total gas flow rate of 40 ml/min

4.4 Effect of the Applied Power

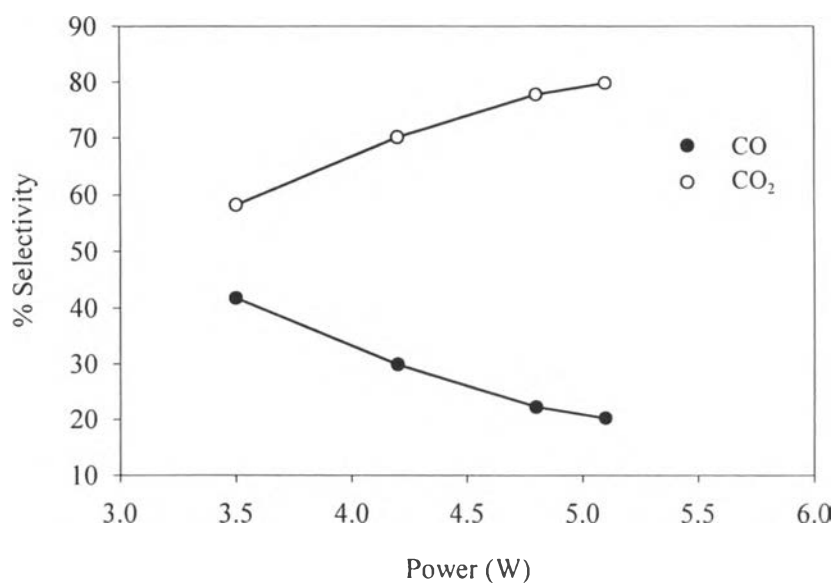### 4.4.1 Effect on the Ethylene and Oxygen Conversion

Figure 4.8 shows the effect of the applied power on the conversion of ethylene and oxygen. The ethylene conversion changes insignificantly with the applied power while a slight increase can be seen for the oxygen conversion. It can be postulated that the higher applied power may result in the higher number of electrons generated, which increase the collision between the electrons and oxygen. Because of the excess amount of oxygen in the feed, the increase in oxygen conversion was also observed. However, there is the optimum power, which is 4.2 W, that results in the highest ethylene and oxygen conversion.

Figure 4.8 Effect of the applied power on the ethylene and oxygen conversion at 200 Hz and the total gas flow of 40 ml/min

4.4.2 Effect on the Product Selectivity

Figure 4.9 shows the effect of the applied power on the product selectivity. In the range of the studied power, 3.5 to 5.1W, the CO_2 selectivity increases from 64.6 to 79.8% and the CO selectivity decreases from 35.4 to 20.2%. The explanation is that when the applied power increases, electrons and O active species are accelerated to higher energy by the electric field. Therefore, the collision between the O active species and CO, which is a high stable molecule in the plasma, becomes more effective.

Figure 4.9 Effect of the applied power on the product selectivity at 200 Hz and the total gas flow rate of 40 ml/min

4.5 Effect of TiO$_2$ Loading

4.5.1 Effect on the Ethylene and Oxygen Conversion

Figure 4.10 illustrates the effect of the TiO$_2$ loading in the plasma reactor. The conversion of ethylene and oxygen do not change with the presence of TiO$_2$ in the plasma reactor and the increase of the applied power.
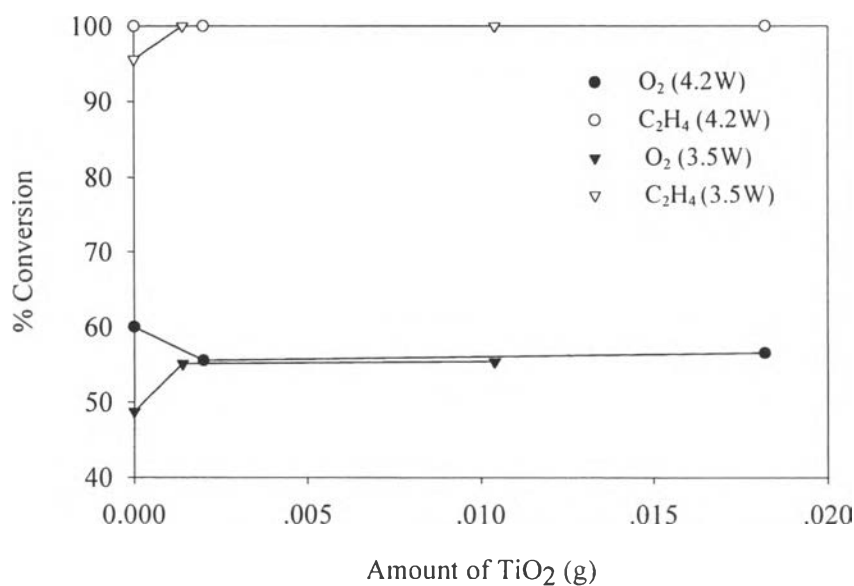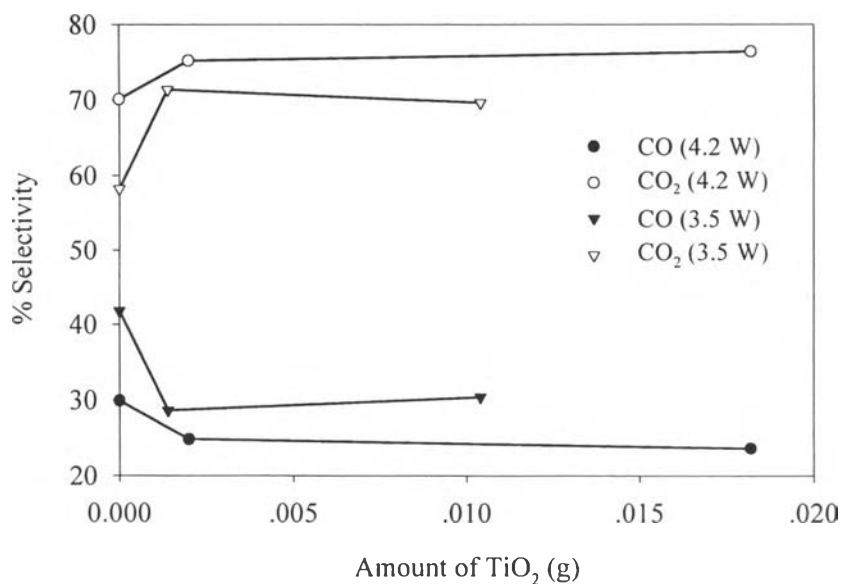
Figure 4.10 Effect of the TiO₂ loading on the ethylene and oxygen conversion at 3.5 and 4.2 W

4.5.2 Effect of TiO₂ Amount on the Product Selectivity

Figure 4.11 shows that the small amount of TiO₂ can increase the CO_2 selectivity from 58 to 71 % at 3.5 W and from 70 to 75 % at 4.2 W, and at the same time it can decrease the CO selectivity. However, further increasing the TiO₂ loading over 0.001 g does not affect both selectivity. The CO_2 selectivity also increases with increasing applied power. The higher applied power possibly generates higher energy species and UV light intensity; therefore, the catalyst can promote more complete oxidation of the ethylene and intermediate products. However, the intensity of the generated UV light may not be high enough to activate all TiO₂ particles so the increase in the TiO₂ content on the support does not affect the product selectivity.

Figure 4.11 Effect of the TiO$_2$ loading on the product selectivity at 3.5 and 4.2 W
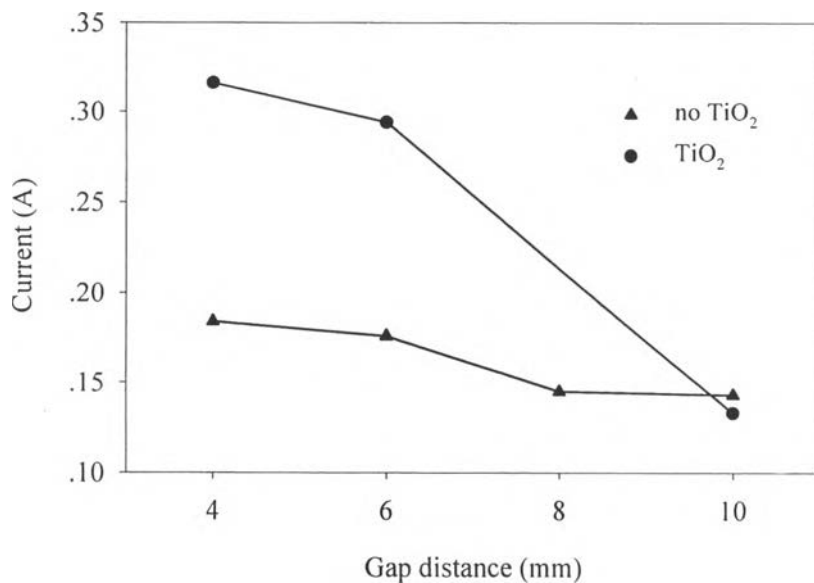
4.6 Effect of the Gap Distance between Electrodes

4.6.1 Effect on the Ethylene and Oxygen Conversion

Results in Figure 4.12 indicate that the conversion of ethylene and oxygen decreases with the decrease in the distance between the electrodes. It can be explained that with a smaller gap, more electrons can move more freely from the wire electrode to the plate electrode with less chance of collision with the reactant gas molecules. On the contrary, in the presence of TiO$_2$, the conversion of ethylene is higher even with the lower electron consumption, which represents as current in Figure 4.13. That is due to some ethylene molecules are oxidized on the TiO$_2$ particles.
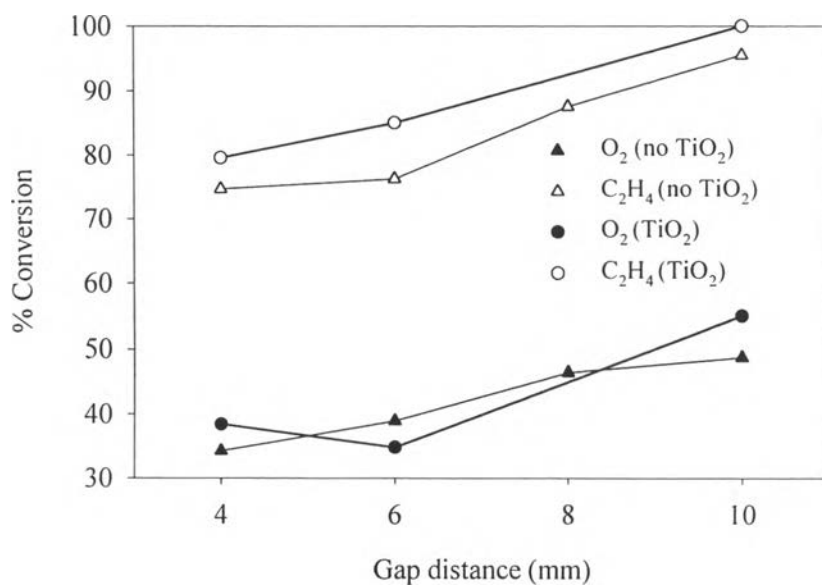
Figure 4.12 Effect of the gap distance on the ethylene and oxygen conversion at 3.5 W, 200 Hz, and the total gas flow rate of 40 ml/min

Figure 4.13 Effect of the gap distance on the current at 3.5 W with and without the TiO_2 coated on glass wool

4.5.1 Effect on the Product Selectivity

The CO_2 selectivity increases with the increase in the gap distance because more radicals such as OH, O, HCO, and HO_2 for oxidizing CO to CO_2 are present (Tsai *et al.*, 2001). The formation of OH groups in the plasma could also prevent the recombination of the positive holes and electrons on the TiO_2 particles; therefore, more ethylene molecules and by-products are adsorbed and oxidized to CO_2 (Peral, 1997).
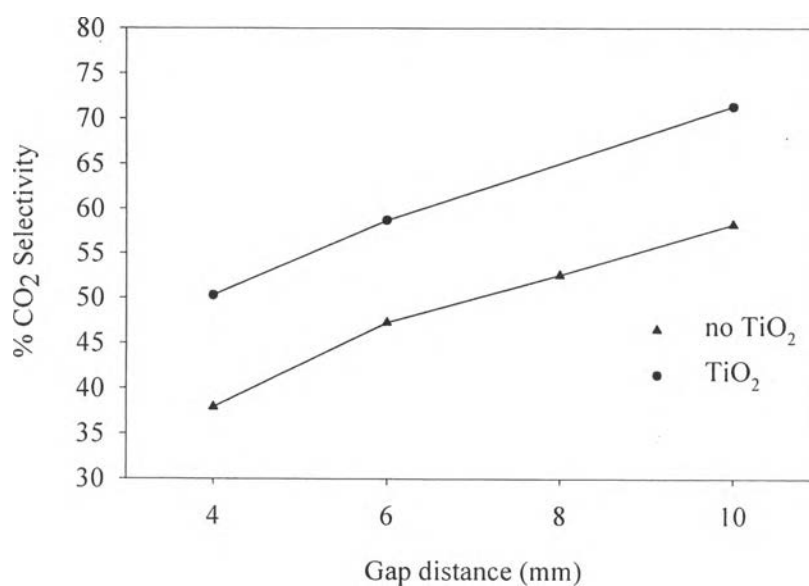
Figure 4.15 Effect of the gap on the CO selectivity with and without TiO_2 at 3.5 W, 200 Hz, and the total gas flow rate of 40 ml/min