

CHAPTER IV

RESULTS AND DISCUSSION

4.1 Au–CuO/CeO₂ Catalysts

Hydrogen production by oxidative steam reforming of methanol (OSRM) was examined over Au-CuO/CeO₂ catalysts prepared by co-precipitation method. The reaction conditions, such as the effect of catalyst pretreatment, Cu/Au (wt/wt) ratio, calcination temperature, H₂O/CH₃OH molar ratio and, O₂/CH₃OH molar ratio were studied in detail.

The catalytic activity tests were carried out in a vertical pyrex glass microreactor by packing with 100 mg catalyst of 80–120 mesh inside. The activity was investigated at varied temperature in the range of 200 to 400°C under atmospheric pressure. The characterization results from several techniques including TPR, FT-IR, UV-vis, XRD, TEM, TPO, AAS, and BET surface area of the prepared catalysts were used to explain the catalytic activity and selectivity of the prepared catalysts in this part.

4.1.1 Effect of Catalysts Pretreatments

To study the effect of catalyst pretreatment on the catalytic performance of 3%wt (1:1), (Cu/Au), Au-CuO/CeO₂ catalyst, the catalysts were pretreated by O₂ and N₂ at 150 °C for 1 hr. The 3%wt (1:1) of bimetallic, Cu/Au, based on weight ratio catalysts prepared by co-precipitation technique with were calcined at 400°C for 4 hours. For the reaction conditions of OSRM, the H₂O/CH₃OH molar ratio and O₂/CH₃OH molar ratio were also initially operated at 2:1 and 0.25:1, respectively. Figure 4.1 shows the methanol conversion and hydrogen yield in the reaction temperature range of 200 to 400°C with different pretreatment gas conditions.

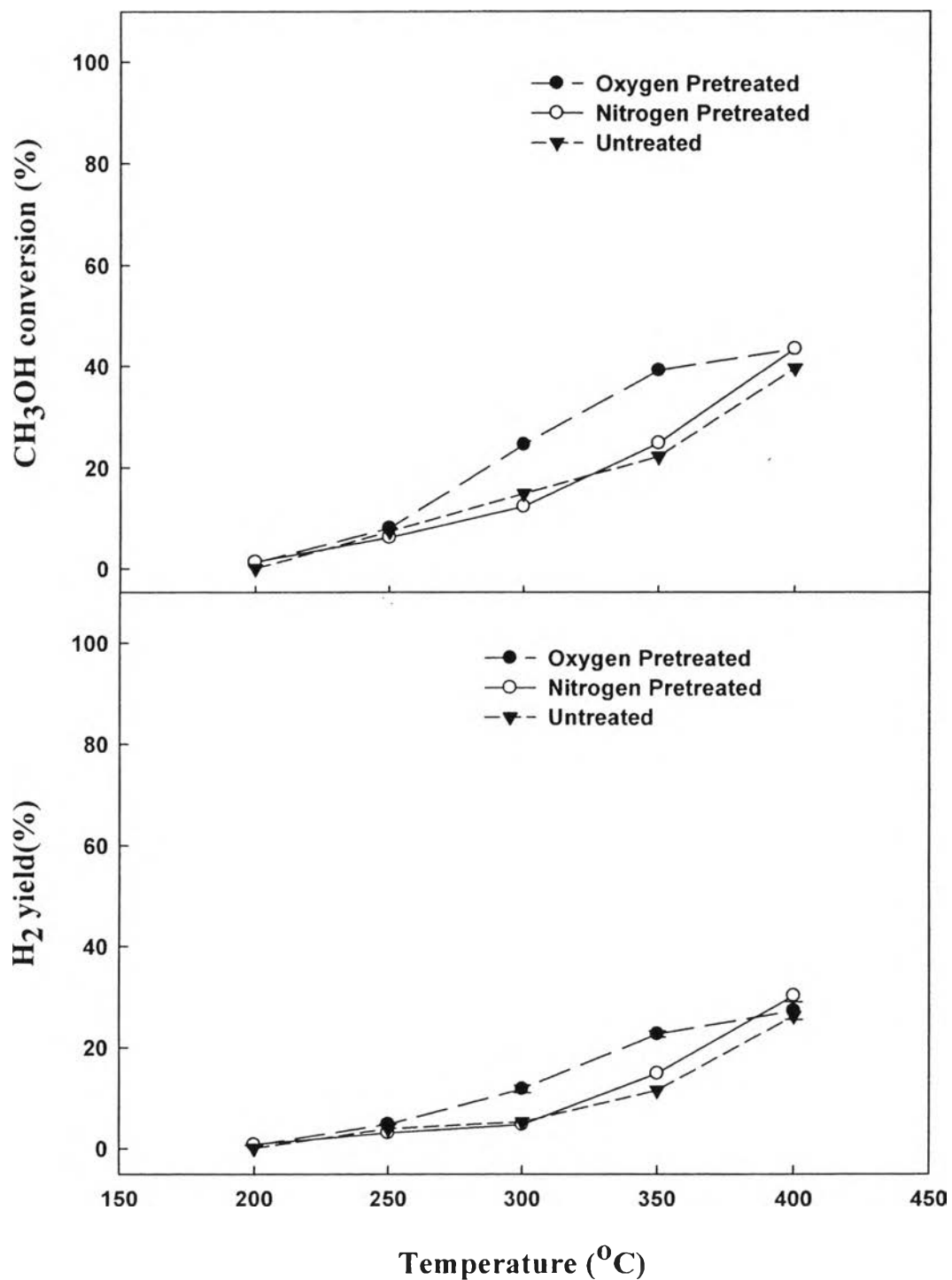


Figure 4.1 Effect of catalyst pretreatment on methanol conversion and hydrogen yield over 3%wt (1:1), (Au/Cu), Au-CuO/CeO₂ catalysts calcined at 400°C (Reaction conditions: O₂/H₂O/CH₃OH molar ratio = 0.63:2:1).

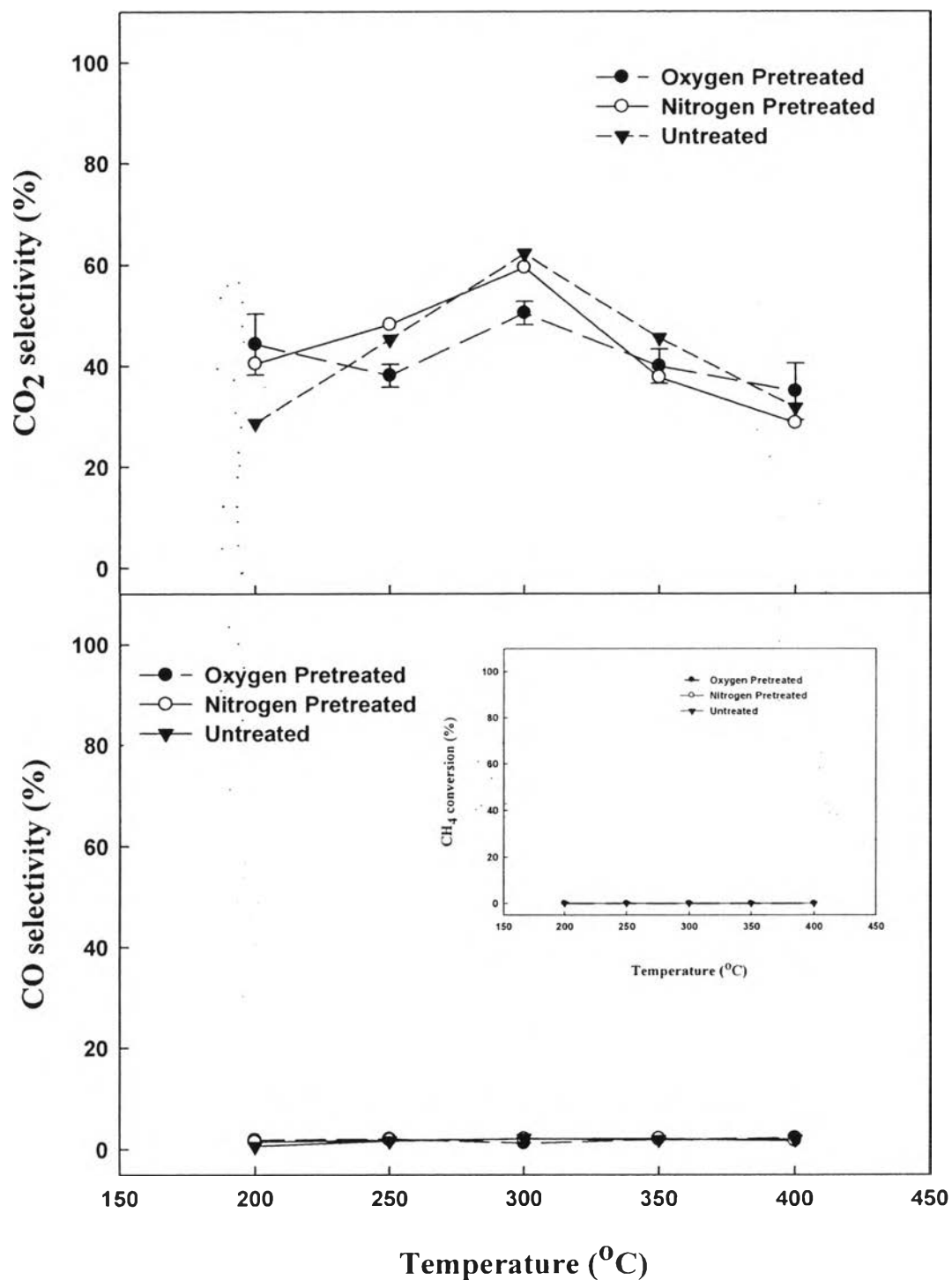


Figure 4.2 Effect of catalyst pretreatment on CO₂, CO, and CH₄ selectivity over 3%wt. (1:1), (Cu/Au), Au-CuO/CeO₂ catalysts calcined at 400°C (Reaction conditions: O₂/H₂O/CH₃OH molar ratio = 0.63:2:1).

It is clearly seen that both methanol conversion and hydrogen yield slightly increased for O₂ pretreatment when compared with N₂ pretreatment. It could be implied that the flowing of O₂ gas might fulfill the oxygen vacancies at the surface of CeO₂ support which plays an important role for improving the catalytic activity in the whole range of reaction temperature (200–400°C). Therefore, O₂ pretreatment was considered as an appropriate condition for the initial study. Tang *et al.*, 1999 was found that Cu species reduction at the Cu-Ce interface play a decisive role for O₂ activation by transferring O₂ from Cu-Ce interface to adsorbed reactant enhance CO oxidation reaction. This also illustrated the small amounts of CO formation during the OSRM reaction, which was still acceptable for PEM fuel cell application.

4.1.1.1 X-Ray Diffraction (XRD)

The effect of catalyst pretreatment on the nature of Au catalysts has been studied by XRD. Figure 4.3 shows the XRD patterns of the catalysts, which consists of O₂ pretreated, N₂ pretreated and untreated. The phase composition obtained from the XRD patterns of all catalysts was very similar.

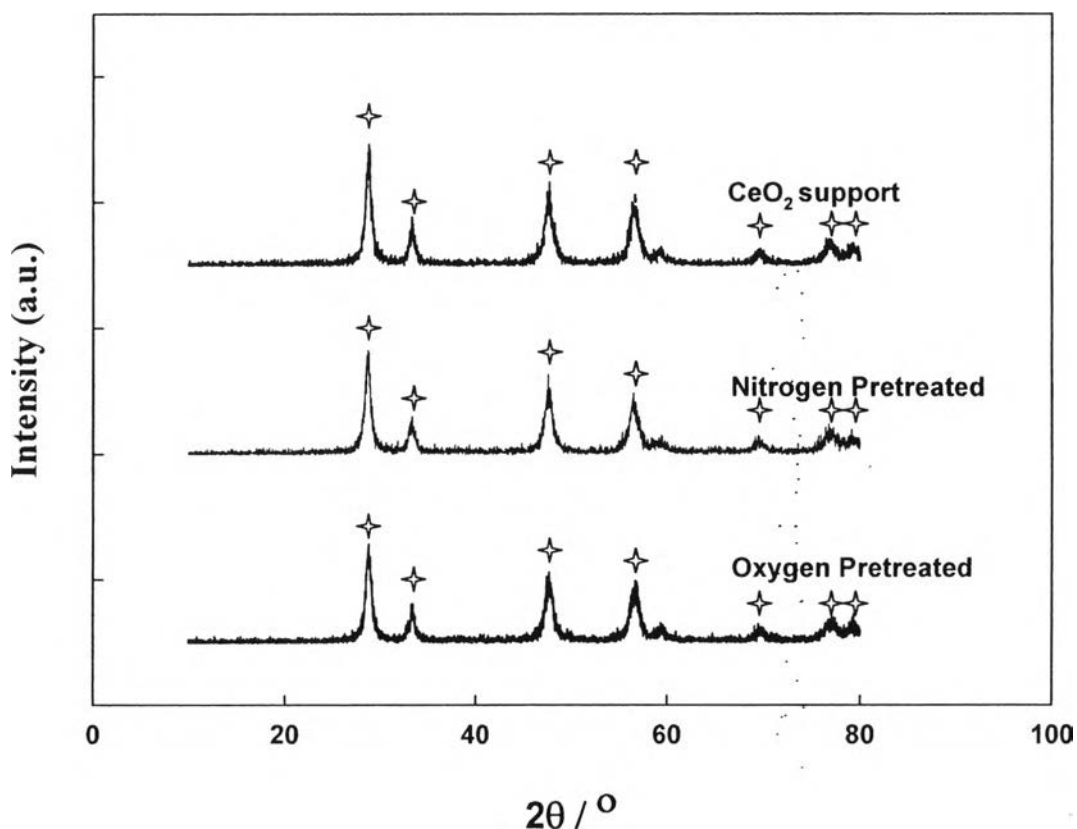


Figure 4.3 XRD patterns of Au-CuO/CeO₂ catalysts with various pretreatment conditions.

4.1.2 Effect of Au/Cu (wt/wt) Ratio on Catalytic Performance

For studying the effect of Cu/Au (wt/wt) ratios, the weight ratios of Cu and Au (Cu/Au) —must be varied whereas the total weight percentage was kept constant at 3%wt over Au-CuO/CeO₂ catalysts. The catalyst were prepared by co-precipitation technique, calcined at 400°C, and then tested for catalytic activity at the reaction condition of H₂O/CH₃OH molar ratios and O₂/CH₃OH molar ratio of 2:1 and 0.63:1, respectively.

Figure 4.4 shows the methanol conversion and hydrogen yield increased with increasing metal weight ratio during the reaction temperature from 200 to 400°C. It can be seen that CuO/CeO₂ is the most active catalyst for this experiment. Nevertheless, bi-metallic catalyst with a 5:1, (Cu/Au) can exhibit the

highest catalytic performance (98% methanol conversion), compared to 1:1 and 1:5, respectively. The H₂ yield for each catalyst was slightly different during reaction temperature range. It can be concluded that doping of Cu on Au-CuO/CeO₂ catalyst can increase the catalytic activity in the OSRM, as shown in Figure 4.5. Au/CeO₂ exhibited the lowest catalytic activity in terms of methanol conversion and H₂ yield.

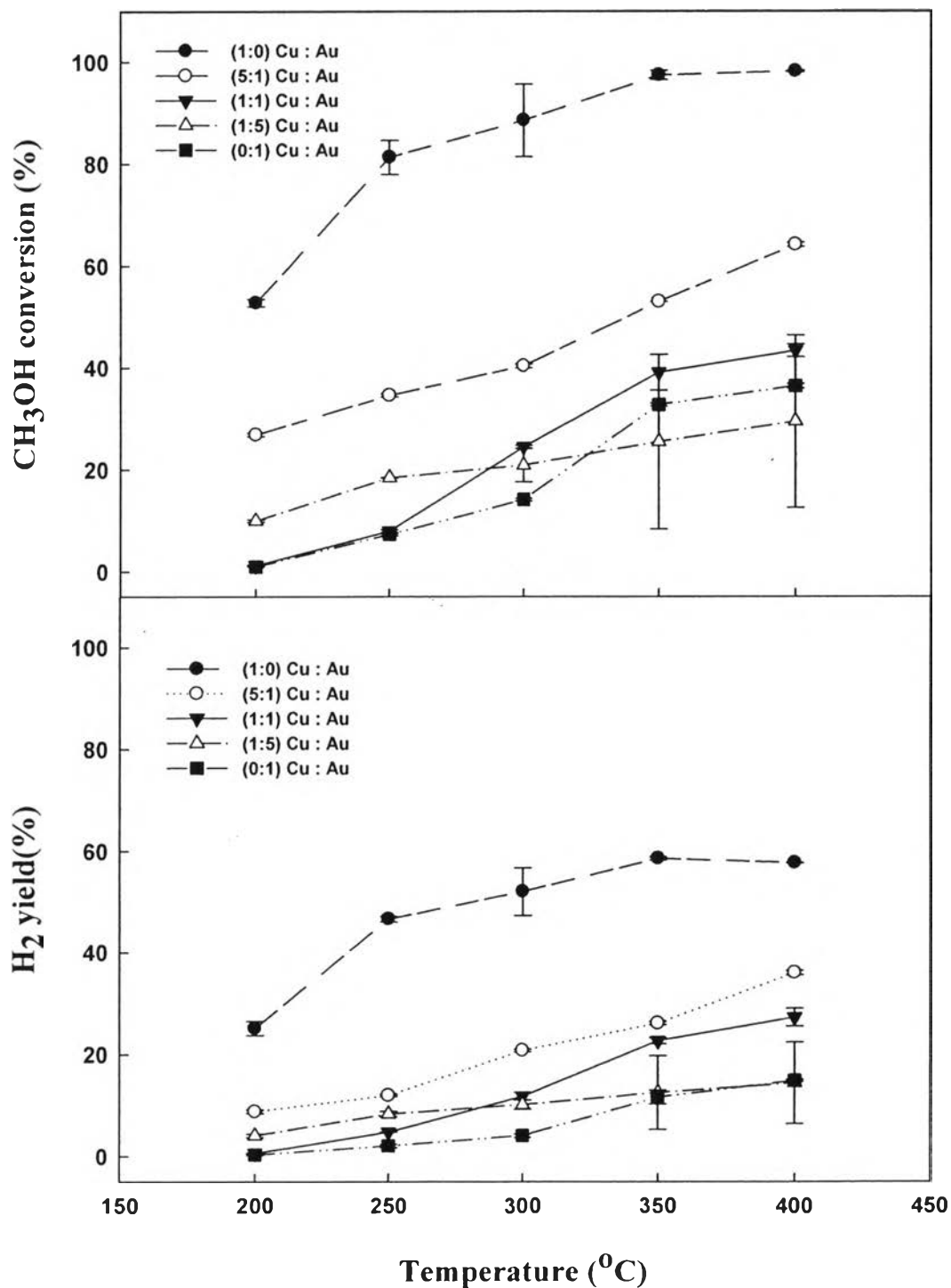


Figure 4.4 Effect of Au/Cu (wt/wt) ratio on the methanol conversion and hydrogen yield over 3%wt of Au-CuO/CeO₂ catalysts calcined at 400°C (Reaction conditions: O₂/H₂O/CH₃OH molar ratio = 0.63:2:1).

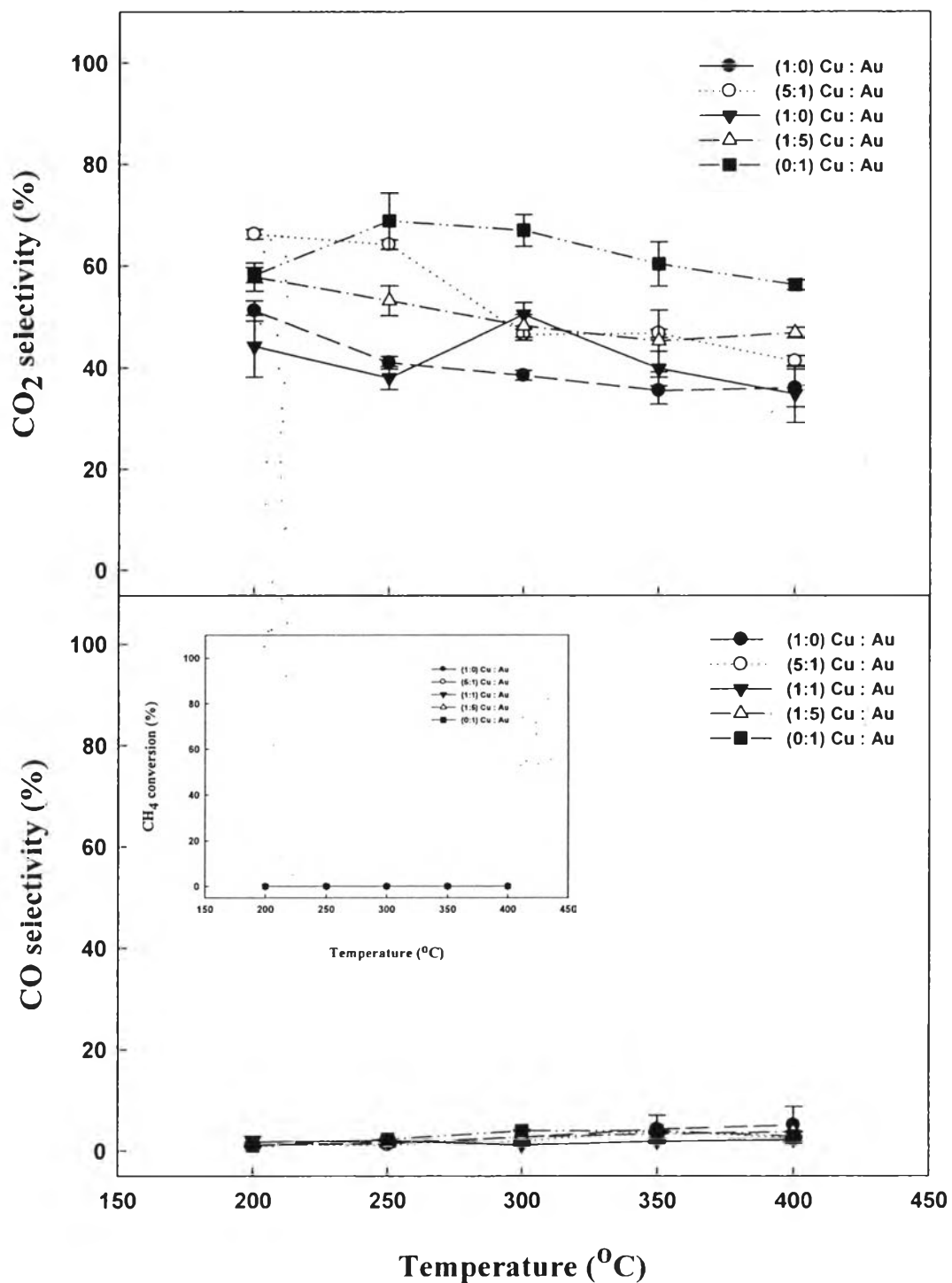


Figure 4.5 Effect Au/Cu (wt/wt) ratio on CO₂, CO, and CH₄ selectivity over 3%wt (1:1), (Cu/Au), Au-CuO/CeO₂ catalysts calcined at 400°C (Reaction conditions: O₂/H₂O/CH₃OH molar ratio = 0.63:2:1).

4.1.2.1 X-Ray Diffraction (XRD)

The XRD patterns of the prepared catalysts shown in Figure 4.6 are very similar to that corresponding to CeO₂. All catalysts presented a strong diffraction peak at $2\theta = 28.5^\circ$, which is the characteristics of fluorite structure of CeO₂. The other peaks at 33.08, 47.47, 56.33, 59.08, 69.40, 76.69, and 79.07° were corresponding to CeO₂ (200), CeO₂ (220), CeO₂ (311), CeO₂ (222), CeO₂ (400), CeO₂ (331), and CeO₂ (420) for CuK α (1.5406 Å) radiation (Arena *et al.*, 2006). Optionally, Au (111) peak can be detected on 0:1, 1:5, and 1:1 of Au-CuO/CeO₂ catalysts which were observed at 2θ of 38.35°. It's agreement that increasing of Au content in Au-CuO/CeO₂ catalyst lead to increasing of Au particle size calculated by Scherrer equation and result summarized in Table 4.1 (Arena *et al.*, 2006). No CuO preaks are observed at $2\theta = 35.7, 39.3, 61.5,$ and 66.5 for all catalysts, probably due to very low metal loading and its high dispersion.

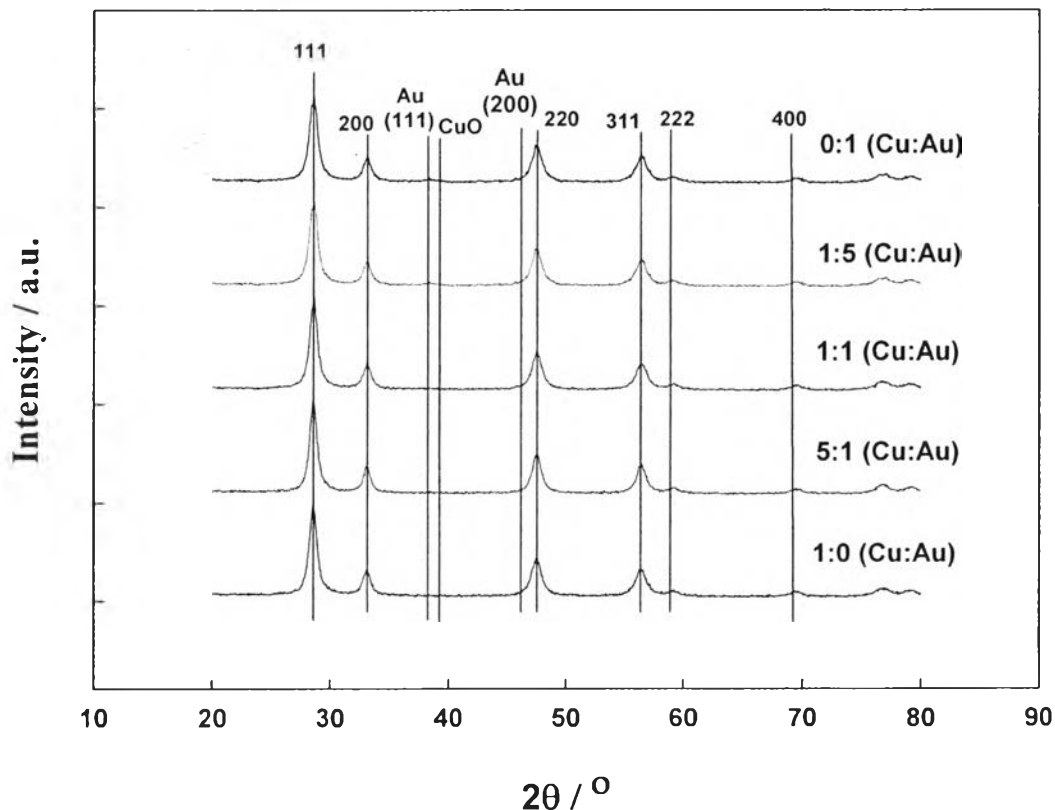


Figure 4.6 XRD patterns of Au-CuO/CeO₂ catalysts with various Cu/Au (wt/wt) ratios.

Table 4.1 Crystallite sizes of the Au-CuO/CeO₂ catalysts with different (Cu/Cu) molar ratios

Catalyst	Crystallite size (nm.)						
	CeO ₂ (111)	CeO ₂ (200)	CeO ₂ (220)	CeO ₂ (331)	Au (111)	Au (220)	CuO (111)
1:0 (Cu/Au)	13.25	14.73	9.08	22.17	< 5	< 5	14.98
5:1 (Cu/Au)	13.24	14.73	11.86	29.01	< 5	< 5	< 5
1:1 (Cu/Au)	12.04	14.74	11.01	23.82	8.84	< 5	< 5
1:5 (Cu/Au)	13.25	13.40	14.03	35.99	10.18	< 5	< 5
0:1 (Cu/Au)	11.20	11.33	12.85	28.98	11.34	< 5	< 5

4.1.2.2 Surface Area Measurement (BET)

BET surface area of 1:0, 5:1, 1:1, 1:5, 0:1 of 3% Au-CuO/CeO₂ and CeO₂ support are summarized in Table 4.2. After loading Au and CuO on CeO₂ support, the surface areas of all catalysts were drastically decreased. The surface area were approximately in the range of 51.65 to 77.61 m²/g for metal loaded on CeO₂ support catalysts. However, trend of surface area still unclear.

Table 4.2 BET surface areas of CeO₂, 1:0, 5:1, 1:1, 1:5 and 0:1 of Au-CuO/CeO₂

Catalyst	BET surface area (m ² /g)
CeO ₂ support	151.39
1:0 (Cu/Au)	61.20
5:1 (Cu/Au)	69.06
1:1 (Cu/Au)	51.65
1:5 (Cu/Au)	70.73
0:1 (Cu/Au)	77.61

4.1.2.3 Temperature-Programmed Reduction (TPR)

The temperature-programmed reduction (TPR) technique was used to study the reduction profiles of the catalysts, as shown in Figure 4.7. For the ceria support, there is one main peaks observed in this experiment. The broader peak was recorded at 500°C corresponding to the surface reduction from CeO₂ to Ce₂O₃ (Jacobs *et.al.*, 2005). The Au/CeO₂ catalyst shows the peak at low temperature of 135 °C, corresponding to the surface reduction from AuO to Au (Chang *et.al.*, 2008) and for CuO/CeO₂ two major peak was detected at 168°C and 191°C (Perez *et.al.*, 2007). Figure 4.7 indicated that the addition of the Cu metal can catalyze the surface reduction process, shifting the peaks to the lower temperature (Ayastuy *et.al.*, 2010). Indeed the addition of Cu could reduce the metal-support interaction between Au and CeO₂ support where the Cu metal interacted with Au metal on surface of catalyst. Moreover, increasing of CuO content leads catalyst for being active in catalytic performance for all temperature range (200–400°C).

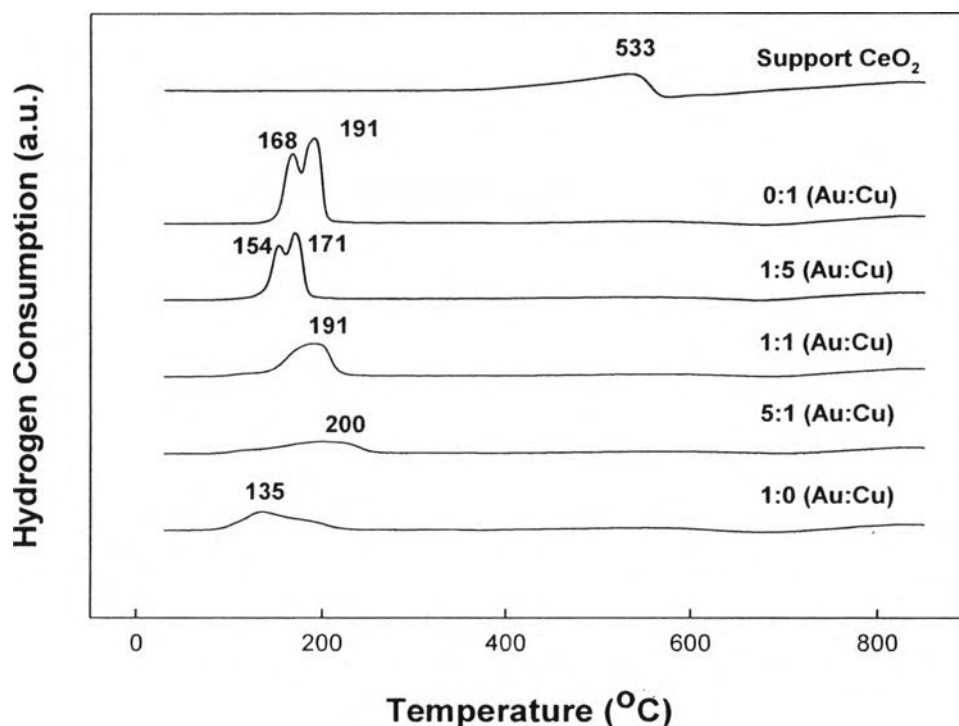


Figure 4.7 TPR profiles of Au-CuO/CeO₂ calcined at 400 °C with different Cu/Au (wt/wt) ratios.

4.1.2.4 UV-visible Spectroscopy

According to stacking of ceria oxide and Au³⁺ band which show in the range of 200–350 nm and < 250 nm (Souza *et al.*, 2008). It leads to undetectable of Au³⁺ on Au-CuO/CeO₂ catalyst by UV spectra. However, the absorption of Au metallic (plasmon) can be detected at 550 nm to confirm catalytic result. Figure 4.8 illustrates that all the catalysts show the absorption from 450-600 nm which referred to gold metallic (plasmon) and it can be seen that an increase of Au content catalyst can pronounce more gold metallic on Au-CuO/CeO₂. Due to the evidence that gold metallic can be more pronounced with increasing of Au content on Au-CuO/CeO₂ catalyst. According to the interfering of CeO₂ band and Au³⁺ band, it can be concluded that state of Au is still unclear.

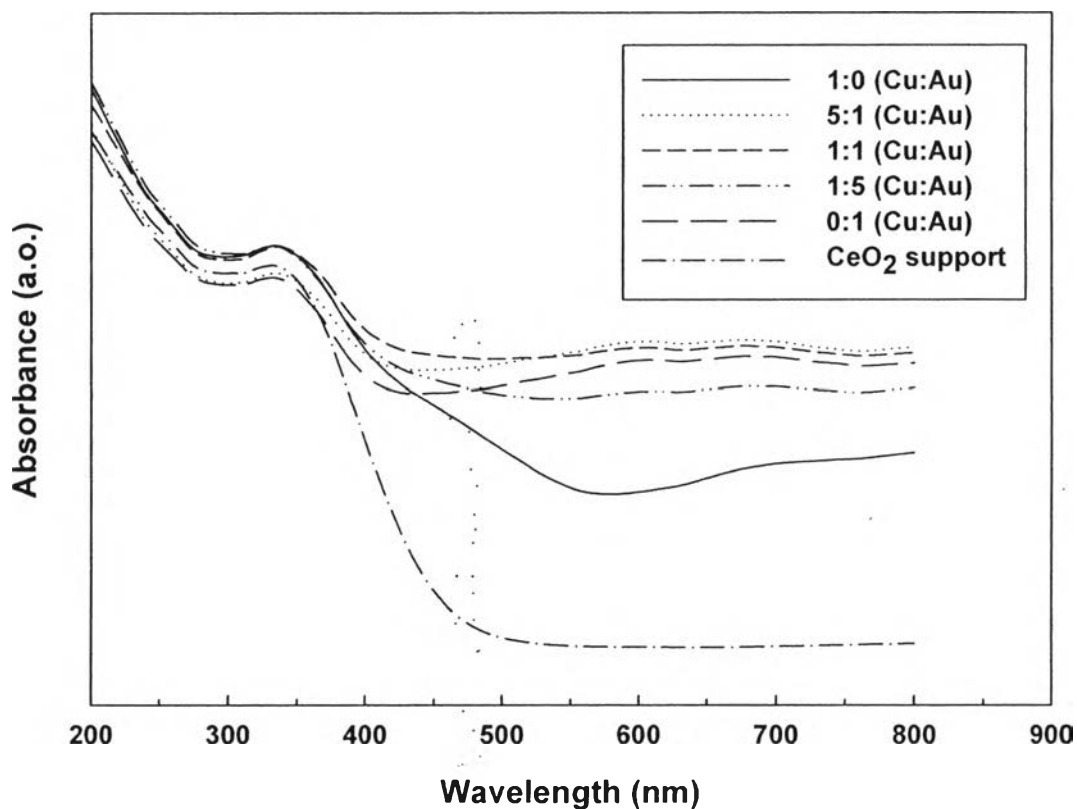


Figure 4.8 Diffuse Reflectance UV-vis spectra of Au species on Au-CuO/CeO₂ with different (Cu/Au) metal ratios.

4.1.3 Effect of Calcination Temperature on Catalytic Performance

To study the effect of calcination temperature on the catalytic performance of 5:1 (Cu/Au) of Au-CuO/CeO₂ catalyst with different calcinations temperatures (200, 300, 400, and 500°C)

It can be seen that both methanol conversion and hydrogen yield were increased for all of the reaction temperature range. The highest catalytic activity was observed by 5:1 of Au-CuO/CeO₂ catalyst calcined at 200°C with methanol conversion at 74.14% and hydrogen yield of 42.96% as shown in Figure 4.9. It was found that the BET surface area decreased with increasing calcination temperature. It can be concluded that the catalytic activity was dropped since the sintering of the catalyst occurred. According to the sintering phenomena of catalyst, TEM was used

to confirm the agglomeration of active metal particles. Figure 4.10 shows that CO, CO₂, and CH₄ selectivity have slightly different for each calcination temperature.

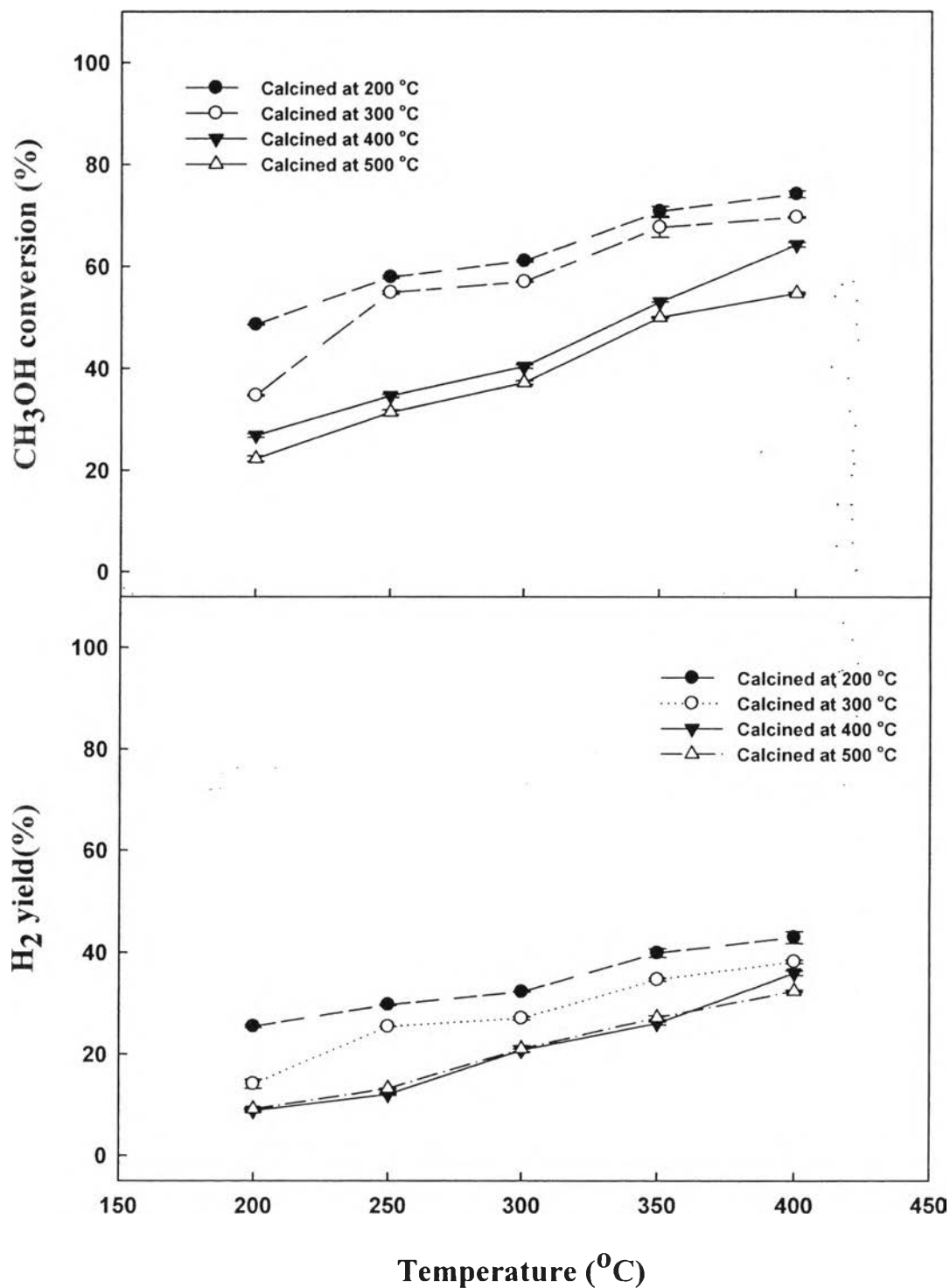


Figure 4.9 Effect of calcination temperature on methanol conversion and hydrogen yield over 3%wt (5:1), (Cu/Au), Au-CuO/CeO₂ catalysts with various calcination temperatures (Reaction conditions: O₂/H₂O/CH₃OH molar ratio = 0.63:2:1).

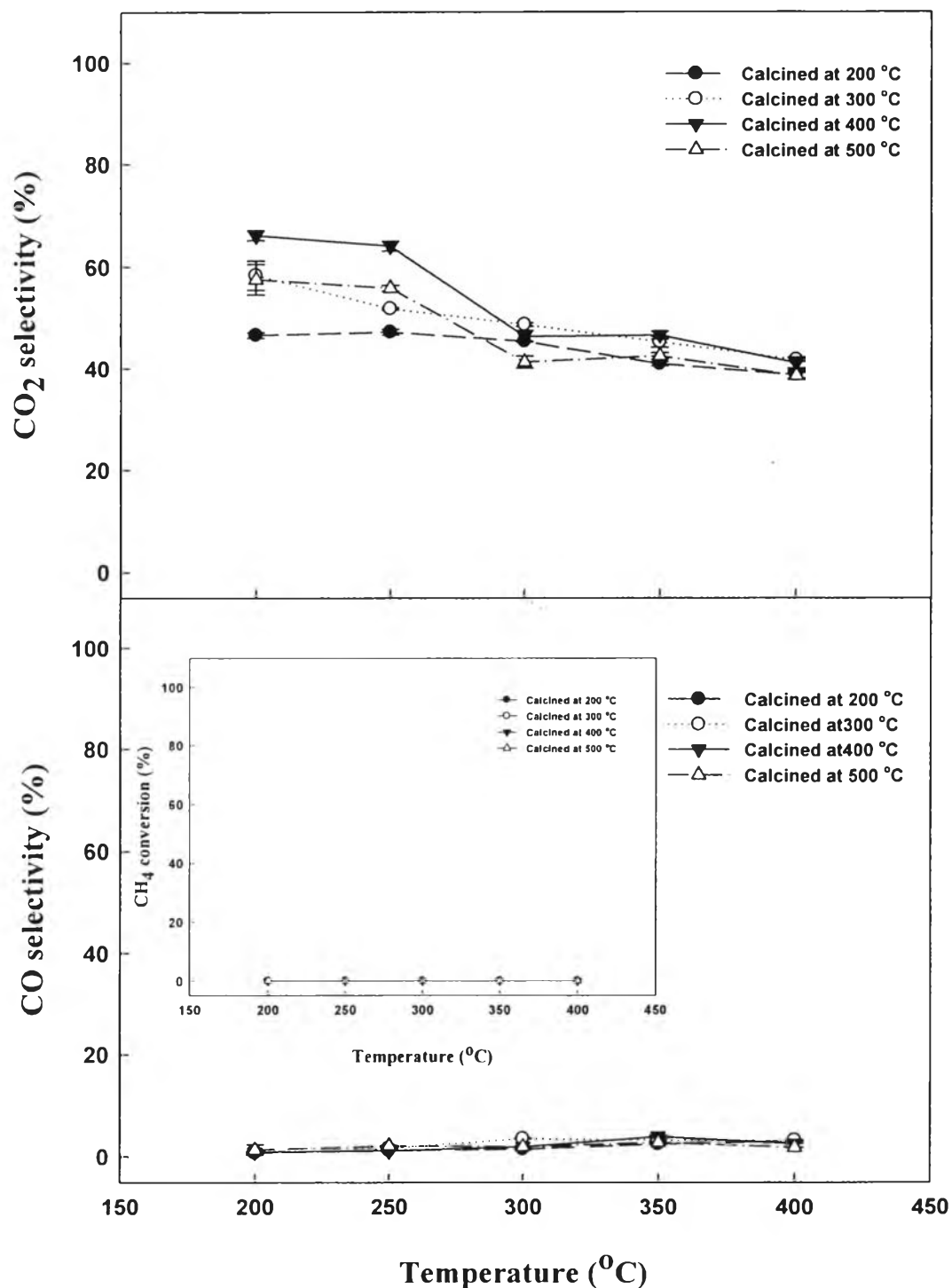


Figure 4.10 Effect of calcination temperature on CO₂, CO, and CH₄ selectivity over 3%wt (5:1), (Cu/Au), Au-CuO/CeO₂ catalysts with various calcination temperatures (Reaction conditions: O₂/H₂O/CH₃OH molar ratio = 0.63:2:1).

4.1.3.1 X-Ray Diffraction (XRD)

From Figure 4.11, the XRD patterns of 5:1 Au-CuO/CeO₂ catalyst with different calcination temperatures show that the gold reflections are not clearly visible for all catalysts. This shows that Au particles are dispersed as small particles on the surface of the support. It could be implied that with low loading of Au and Cu on catalyst would be under detectable capability of the machine (5nm), as shown in Table 4.3.

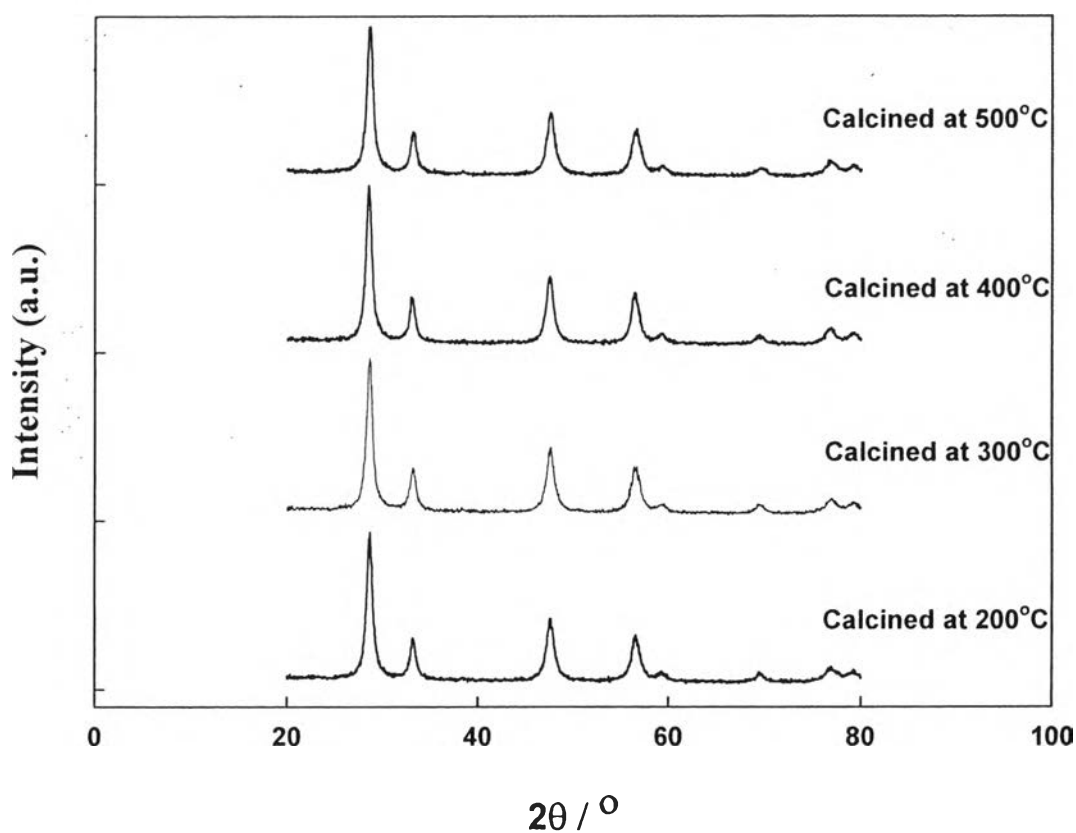


Figure 4.11 XRD patterns of 3%wt (5:1),(Cu:Au) of Au-CuO/CeO₂ catalysts with different calcination temperatures.

Table 4.3 Crystallite sizes of the Au-CuO/CeO₂ catalyst with different calcination temperatures

Catalyst	Crystallite size (nm.)						
	CeO ₂ (111)	CeO ₂ (200)	CeO ₂ (220)	CeO ₂ (331)	Au (111)	Au (220)	CuO (111)
Calcined at 200°C	13.25	16.38	17.16	29.99	< 5	< 5	< 5
Calcined at 300°C	13.25	13.39	12.86	30.15	< 5	< 5	< 5
Calcined at 400°C	13.24	14.73	11.86	29.01	< 5	< 5	< 5
Calcined at 500°C	13.25	13.40	14.03	35.99	< 5	< 5	< 5

4.1.3.2 Surface Area Measurement (BET)

Catalyst calcinations is generally believed to be a necessary step of catalyst activation. The surface areas of 3% 5:1,(Cu/Au) of Au-CuO/CeO₂ catalyst with various calcination temperatures are summarized in Table 4.4. The surface area of catalysts were decreasing with increasing of calcination temperature from 98.43 m²/g (200°C) to 62.52 m²/g (500°C). High calcination temperature strongly affected the sintering of gold cluster which lead to the blocking of surface area of catalyst by large metal particles. This observation agrees with those of (Eun *et al.*, 1999), they also observed a decrease in surface area with increasing calcinations temperature.

Table 4.4 BET surface areas of 3% 5:1,(Cu/Au) of Au-CuO/CeO₂ catalyst different calcinations temperature

Catalyst	BET surface area (m ² /g)
CeO ₂ support	151.39
Calcined at 200°C	98.43
Calcined at 300°C	80.61
Calcined at 400°C	69.06
Calcined at 500°C	62.52

4.1.3.3 Temperature-Programmed Reduction (TPR)

From Figure 4.12, it is clearly seen that increasing of calcination temperature did not affect much to interaction of Au and CeO₂ support in Au-CuO/CeO₂ catalyst. Because TPR profiles show the reduction temperature in range of 150 to 182°C. It can be noticed from the TPR profiles that the peak width and the intensity of the first peak at 138 to 159°C decrease with the increase of calcinations temperature. To explain the catalytic activity in the effect of calcinations temperature, the other characterization results could be required to explain this phenomenon.

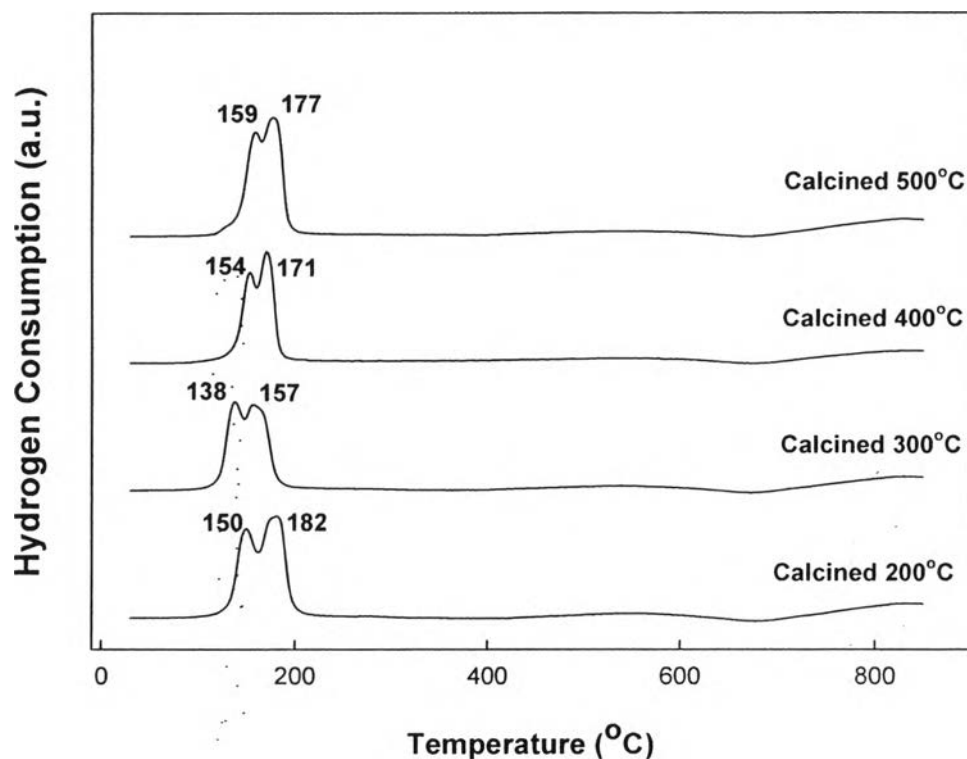


Figure 4.12 TPR profiles of 3%wt (5:1),(Cu:Au) of Au-CuO/CeO₂ different calcination temperatures.

4.1.3.4 UV-visible Spectroscopy

In order to explain the catalytic activity in Figure 4.9, 5:1 of Au-CuO/CeO₂ calcined at 200°C which exhibit the highest catalytic activity for both methanol conversion and hydrogen yield. The Kubelka-Munk transformed UV-vis diffuse reflectance spectra of the samples are shown in Figure 4.13. CeO₂ support shows as absorption band at wavelength below 400 nm and CuO typically shows two adsorption peaks at 247 and 312 nm, which are related to O²⁻ → Cu²⁺ charge transfer transition (Praliaud *et al.*,1998). According to the spectra band in the range of 450-700 nm, gold metallic particle was observed and related to 5:1 of Au-CuO/CeO₂ calcined at 200°C which had the highest amount of Au⁰ species. Moreover, catalysts were fixed the quantity of gold loading at 5:1 (Cu:Au) that linked to the result in Figure 4.13 and it can be observed that Au-CuO/CeO₂ calcined at 200°C showed the

highest crystallinity of Au particle and highest quantity of gold metallic particle opposed with some literature. In 2006 Smolentseva E and coworkers found that the increasing of calcination temperature leads to the increasing of gold metallic particle while our experiment were not the same trend (Smolentseva E *et al.*,2006). On the other hand Tuzovskaya and coworkers reported that gold metallic particle is responsible for high temperature activity. It could be accept that why the catalyst calcined at 200°C yield the highest catalytic activity. However, the 5:1 of Au-CuO/CeO₂ calcined at 500° exhibited a little bit phenomena of sintering. It could be explained Au⁰ play as an important role with catalytic performance. Nevertheless, Bera and Hegde found that the catalytic activity of CO oxidation can be dropped by the increasing of Au³⁺ but, the overlap of Au³⁺ and CeO₂ spectra band made the investigation of Au³⁺ was still unclear. Some literatures reported that changing form of Au³⁺ to Au⁰ is occurring by increasing of calcinations temperature. However, to clearly the cause of agglomeration of gold nanoparticle TEM was used to investigate.

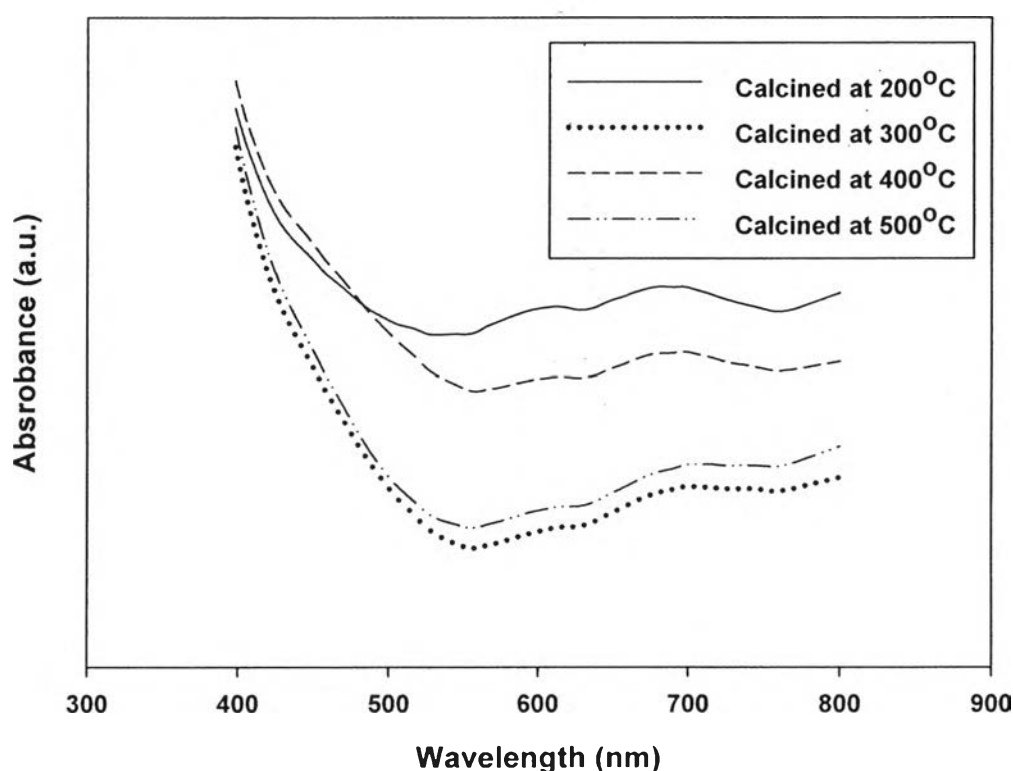
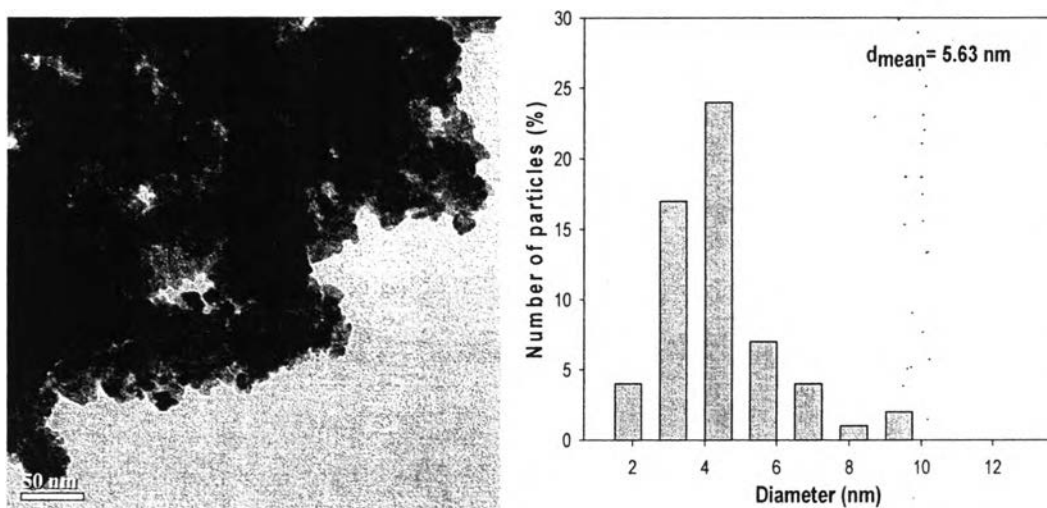


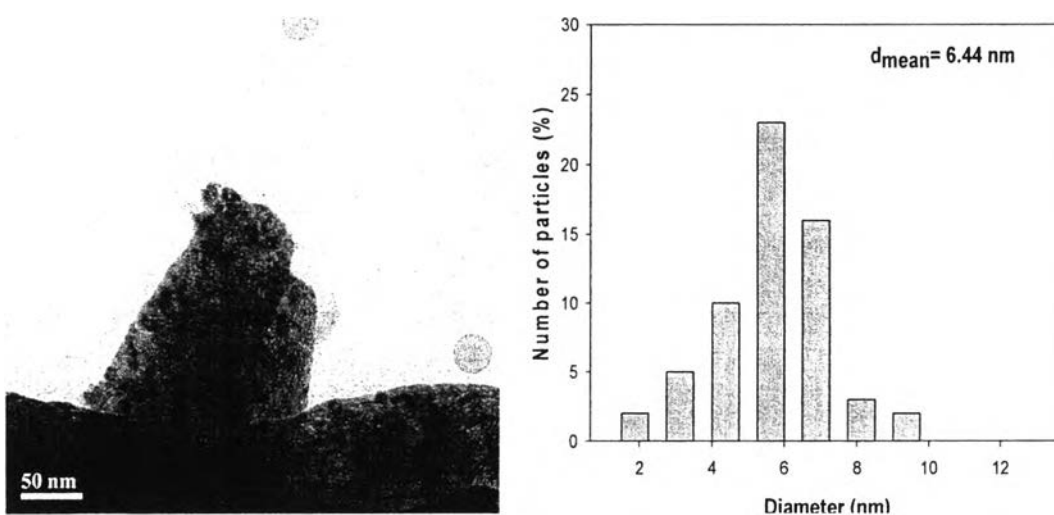
Figure 4.13 Diffuse Reflectance UV-vis spectra of Au species on Au-CuO/CeO₂ with different calcination temperatures.

4.1.3.5 Transmission Electron Microscope (TEM)

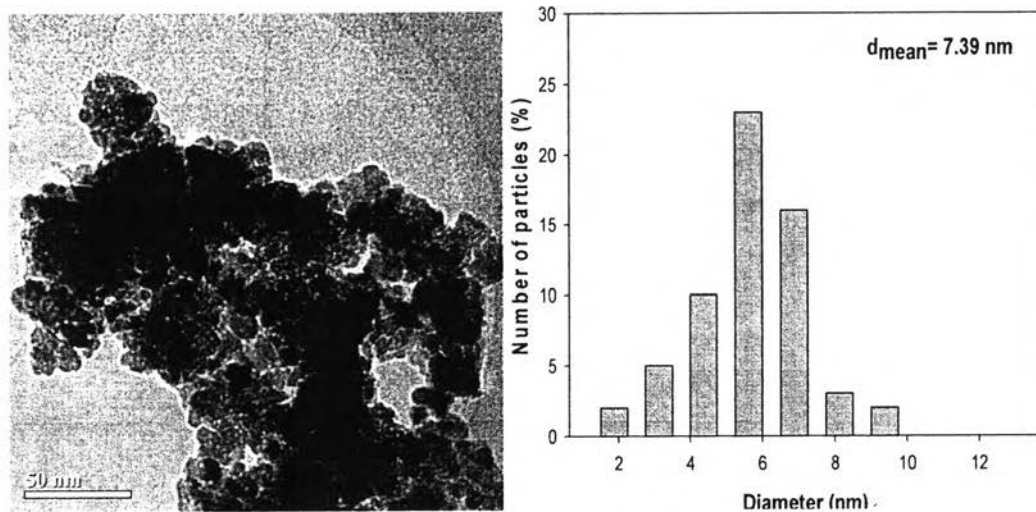
Figure 4.14 shows the TEM images of 5:1 of Au-CuO/CeO₂ with different calcinations temperatures and the average particle size of the catalyst calcined at 200, 300, 400, and 500°C are 5.64 nm, 6.44 nm, 7.39 nm, and 10.16 nm, respectively. The largest gold particle size was observed on the catalyst calcined at 500°C which agrees with BET surface area result that an increase of gold particle would block active surface area on the catalyst surface.



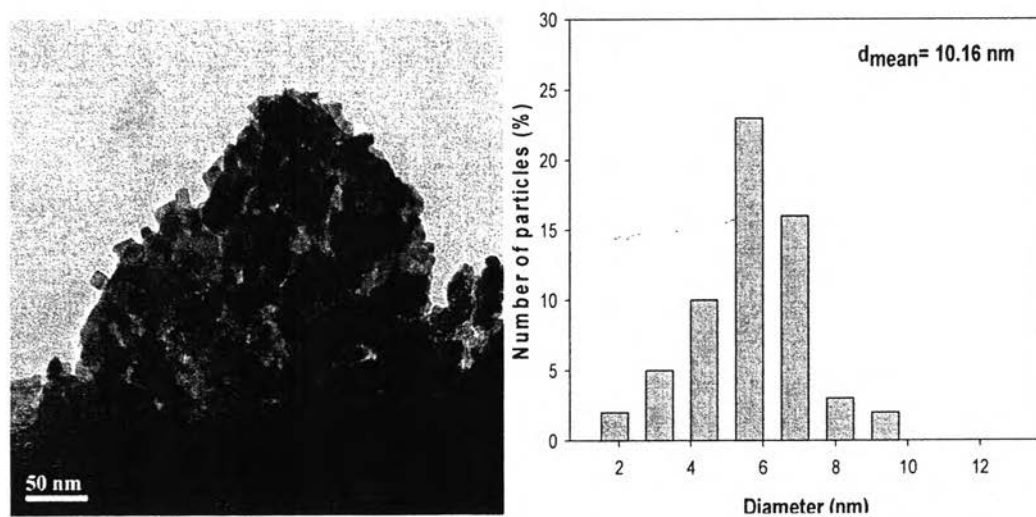
a) Calcined at 200°C



b) Calcined at 300°C



c) Calcined at 400°C



d) Calcined at 500°C

Figure 4.14 TEM images and particle size distribution of Au-CuO/CeO₂ catalysts with various calcination temperatures.

4.1.4 Effect of H₂O/CH₃OH Molar Ratio on the Catalytic Performance of 3% (5:1), (Cu:Au) of Au-CuO/CeO₂

The effect of H₂O/CH₃OH molar ratio on the catalytic activity of 5:1, (Cu/Au) of Au-CuO/CeO₂ calcined at 200°C was studied at the reaction temperature range of 200 to 400°C. The H₂O/CH₃OH molar ratios were studied at 1/1, 2/1, 3/1, and 4/1, respectively.

It is clearly seen that increasing of reaction temperature leads to an increase catalytic activity for both methanol conversion and hydrogen yield. The appropriate condition is 1/1 which can exhibit highest catalytic activity (methanol conversion at 79.49% and hydrogen yield at 51.89%) compared to the other ratios. In order to explain the effect of steam/methanol it refer to forming of hydroxyl group and carbonate species which blocked active sites of catalysts during the reaction (Tabakova *et al.*, 2003). Nevertheless, El-Moemen and co-woker reported that appropriate steam can decompose the carbonate species and catalyst turn to reactive again.

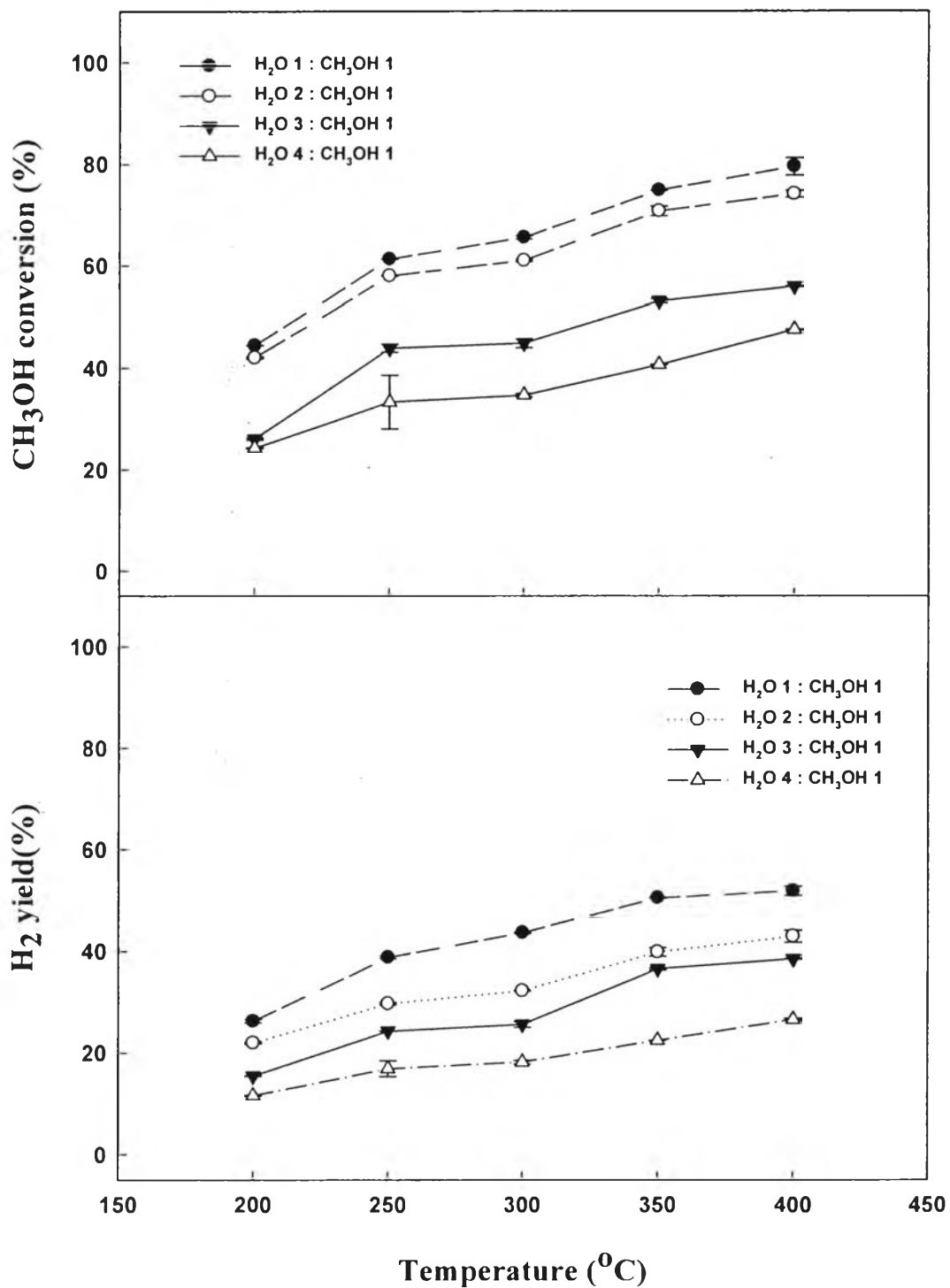


Figure 4.15 Effect H₂O/CH₃OH molar on the methanol conversion and hydrogen yield over 3%wt (5:1) Au-CuO/CeO₂ calcined at 200°C.

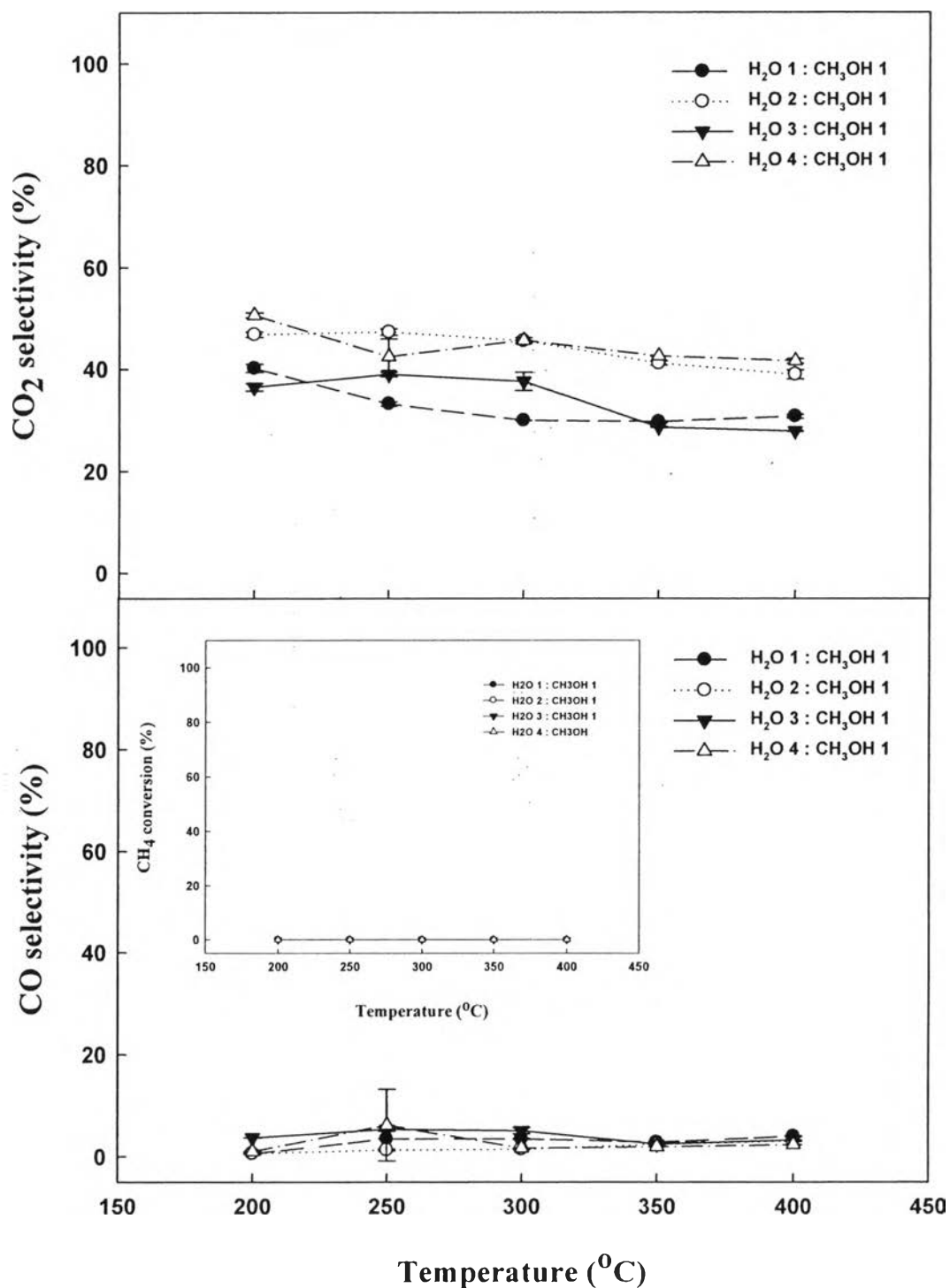


Figure 4.16 Effect H₂O/CH₃OH molar on CO₂, CO, and CH₄ selectivity over 3%wt (5:1) Au-CuO/CeO₂ calcined at 200°C.

4.1.3.1 Fourier Transform Spectroscopy (FT-IR)

The FT-IR results of the spent 5:1, (Cu/Au) of Au-CuO/CeO₂ catalysts with different H₂O/CH₃OH molar ratios are shown in Figures 4.18 and 4.19 for both hydroxyl groups and carbonate species, respectively. For each reaction condition, hydroxyl groups can be formed in the range of 3200-3600cm⁻¹ due to different H₂O/CH₃OH molar ratios. In contrast, carbonate species were detected in the range of 800-1800cm⁻¹ and 2500-3000 cm⁻¹. The different position of each peak indicated different kind carbonate and formate species, as shown in Table 4.5. From Figure 4.18, strong band appear at 2921, 2844, 2853, 2918, and 2847 related to Ce³⁺ and Ce⁴⁺, respectively. Due to feed high amount of steam can lead to WGS reaction which is the side reaction of SRM thus can form formate species as the intermediates (El-Moemen *et al.*, 2008) Nevertheless, high amount of water can easily reduce the carbonate species with decomposition of carbonate species them the catalyst was heated (El-Moemen *et al.*, 2008). It could be implied that the catalytic performance might depend on the formate species stronger than carbonate species due to ability to decompose of carbonate species.

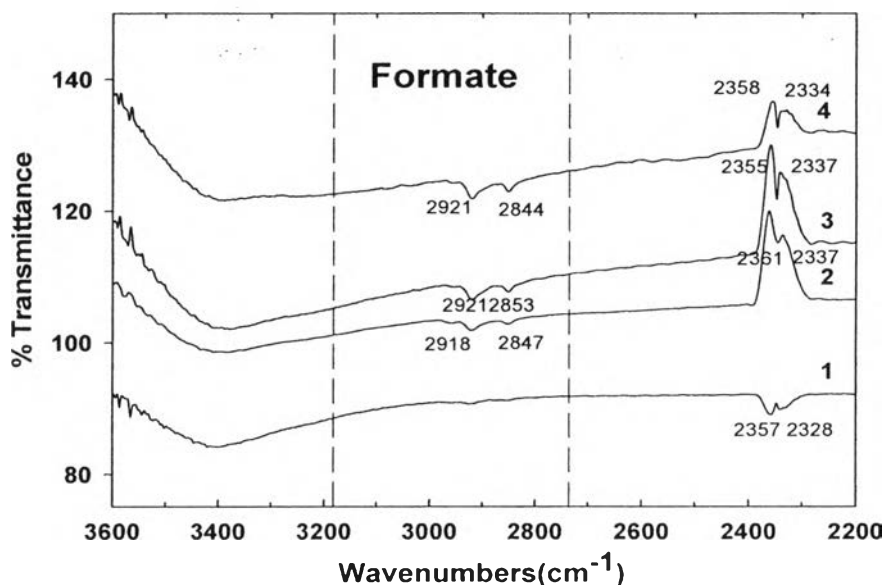


Figure 4.17 FTIR spectra of 3%wt (5:1) Au-CuO/CeO₂ catalysts calcined at 200°C (1) after reaction at H₂O/CH₃OH molar ratio of 1/1; (2) after reaction at H₂O/CH₃OH molar ratio of 2/1; (3) after reaction at H₂O/CH₃OH molar ratio of 3/1; (4) after reaction at H₂O/CH₃OH molar ratio of 4/1.

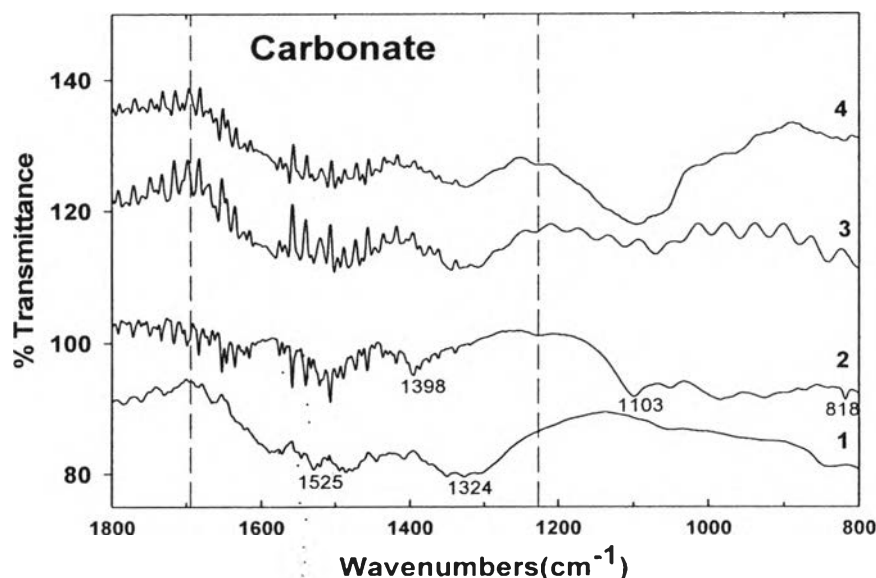


Figure 4.18 FTIR spectra of 3%wt (5:1) Au-CuO/CeO₂ catalysts calcined at 200°C (1) after reaction at H₂O/CH₃OH molar ratio of 1/1; (2) after reaction at H₂O/CH₃OH molar ratio of 2/1; (3) after reaction at H₂O/CH₃OH molar ratio of 3/1; (4) after reaction at H₂O/CH₃OH molar ratio of 4/1.

Table 4.5 Frequency and assignment of carbonate, formate, and intermediate bands on spent of 3%wt Au-CuO/CeO₂ calcined at 200°C (Tabakova *et al.*, 2003)

Wavenumber (cm ⁻¹)	Assignment
2853,1398	Formate species on Ce ⁺
2921, 2844, 2921, 2918, 2847	Formate species on Ce ⁴⁺
818, 2358	Carbonate species on Ceria
1525	Bicarbonate species on Ceria
2334, 2355, 2337, 2331, 2357,2328	Linear CO ₂ weakly interacting ceria

4.1.5 Effect of O₂/CH₃OH Molar Ratio on the Catalytic Performance of 3% (5:1). (Cu: Au) of Au-CuO/CeO₂

To investigate the oxidative steam reforming reaction over 5:1, (Cu/Au) of Au-CuO/CeO₂ calcined at 200°C, the O₂/CH₃OH molar ratios were

varied from 0 to 0.48, 0.96, 1.44, and 1.92, while using the optimum condition of the $\text{H}_2\text{O}/\text{CH}_3\text{OH}$ molar ratio at 1/1 constant. Figure 4.20 showed that the methanol conversion extremely enhance for whole reaction range. It was clearly seen that an increase of O_2 in feedsteam can lead reaction to oxidative steam reforming resulting in an increase on catalytic activity methanol conversion and hydrogen yield in Figure 4.19. At a 0.96 exhibit the highest methanol conversion of ~90% and hydrogen yield of ~51%. Nevertheless, the excess O_2 in feedsteam can shift the reaction to POM resulting in decrease of hydrogen yield. The result in Figure 4.21 showed that the addition of oxygen decreased the hydrogen selectivity than the absence of oxygen which preferred to SRM. The more oxygen contents were added into the reaction, the more reactions preferred to POM which is not favorable at high temperature due to its exothermic reaction.

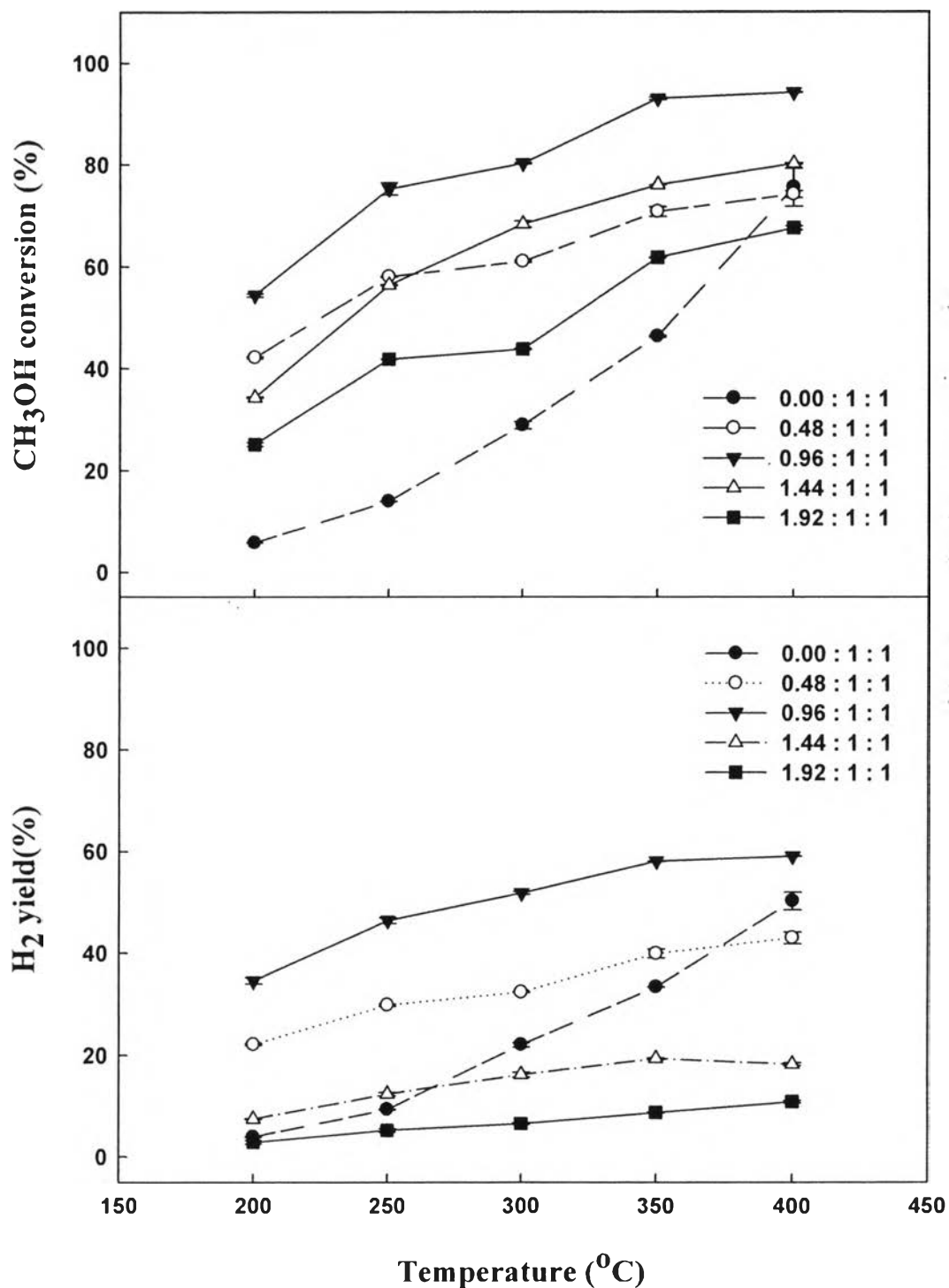


Figure 4.19 Effect O₂/CH₃OH molar ratio on the methanol conversion and hydrogen yield over 3%wt (5:1) Au-CuO/CeO₂ calcined at 200°C.

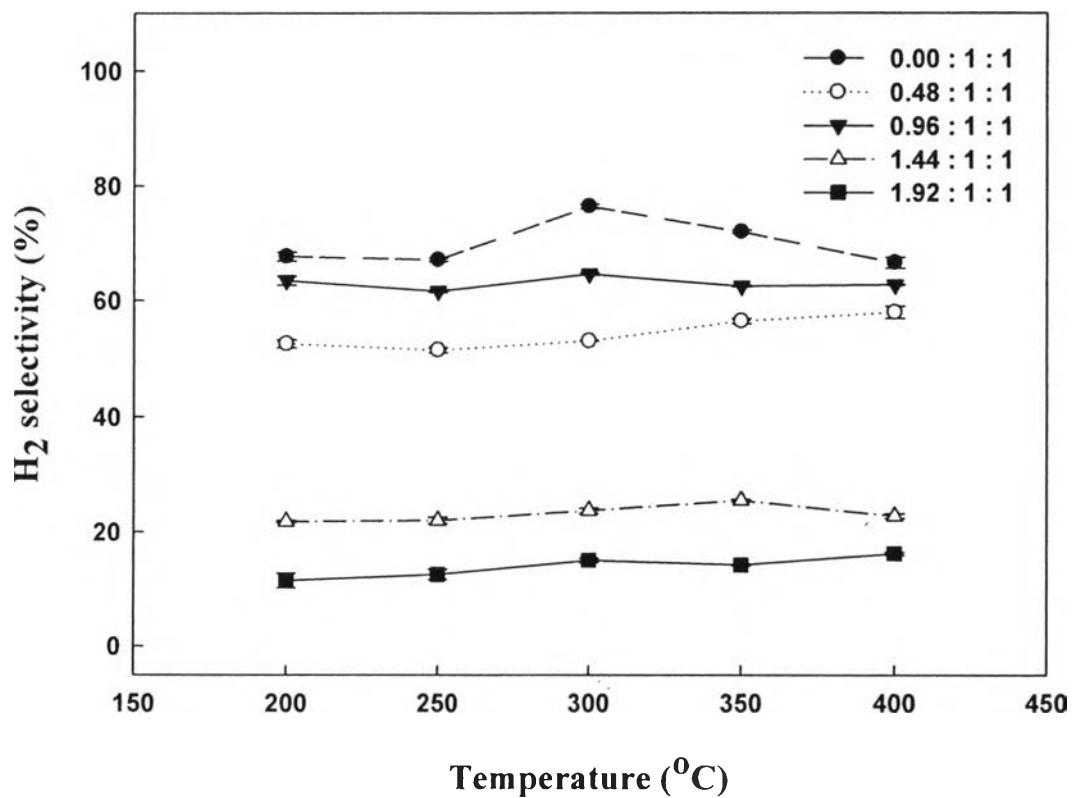


Figure 4.20 Effect O₂/CH₃OH molar ratio on H₂ selectivity over 3%wt (5:1) Au-CuO/CeO₂ calcined at 200°C.

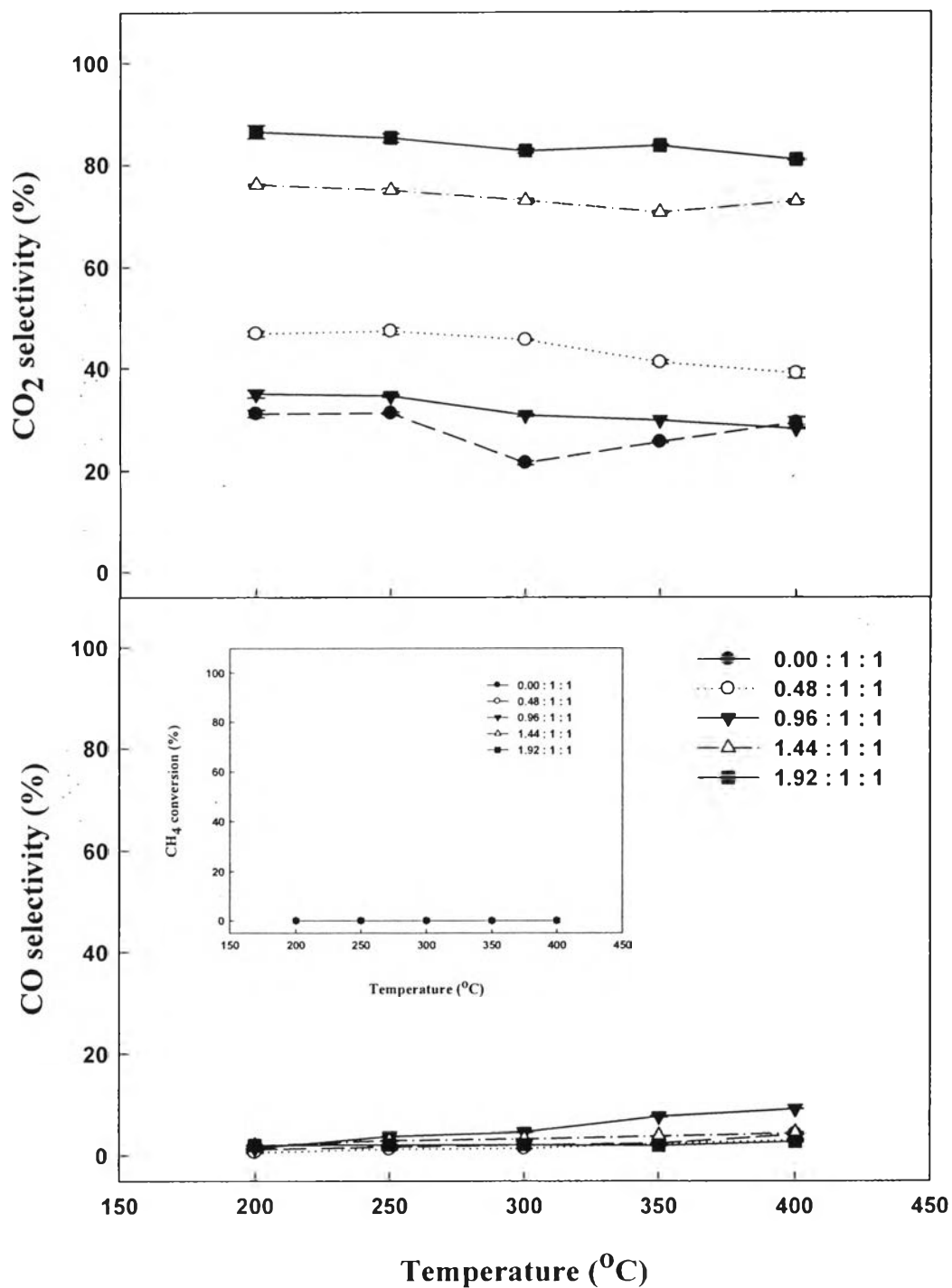


Figure 4.21 Effect O_2/CH_3OH molar on CO_2 , CO , and CH_4 selectivity over 3%wt (5:1) Au-CuO/CeO₂ calcined at 200°C.

4.1.6 Comparison of Catalytic Performance between Support and of 3% (5:1), (Cu: Au) of Au-CuO/CeO₂ calcined at 200°C

To evaluate the catalytic behavior of the prepared catalysts with and without metal loading, the 3%wt 5:1 of Au-CuO/CeO₂ and CeO₂ catalysts calcined at 400°C were compared for the OSRM reaction at the O₂/H₂O/CH₃OH molar ratio at 0.96/1/1 in the temperature range of 200 to 300°C

As illustrated in Figure 4.23, the gap differences in methanol conversion and hydrogen yield between Au-Cu)/CeO₂ and CeO₂ were large. As expected, Au-CuO/CeO₂ response for high catalytic activity in the whole range of reaction.

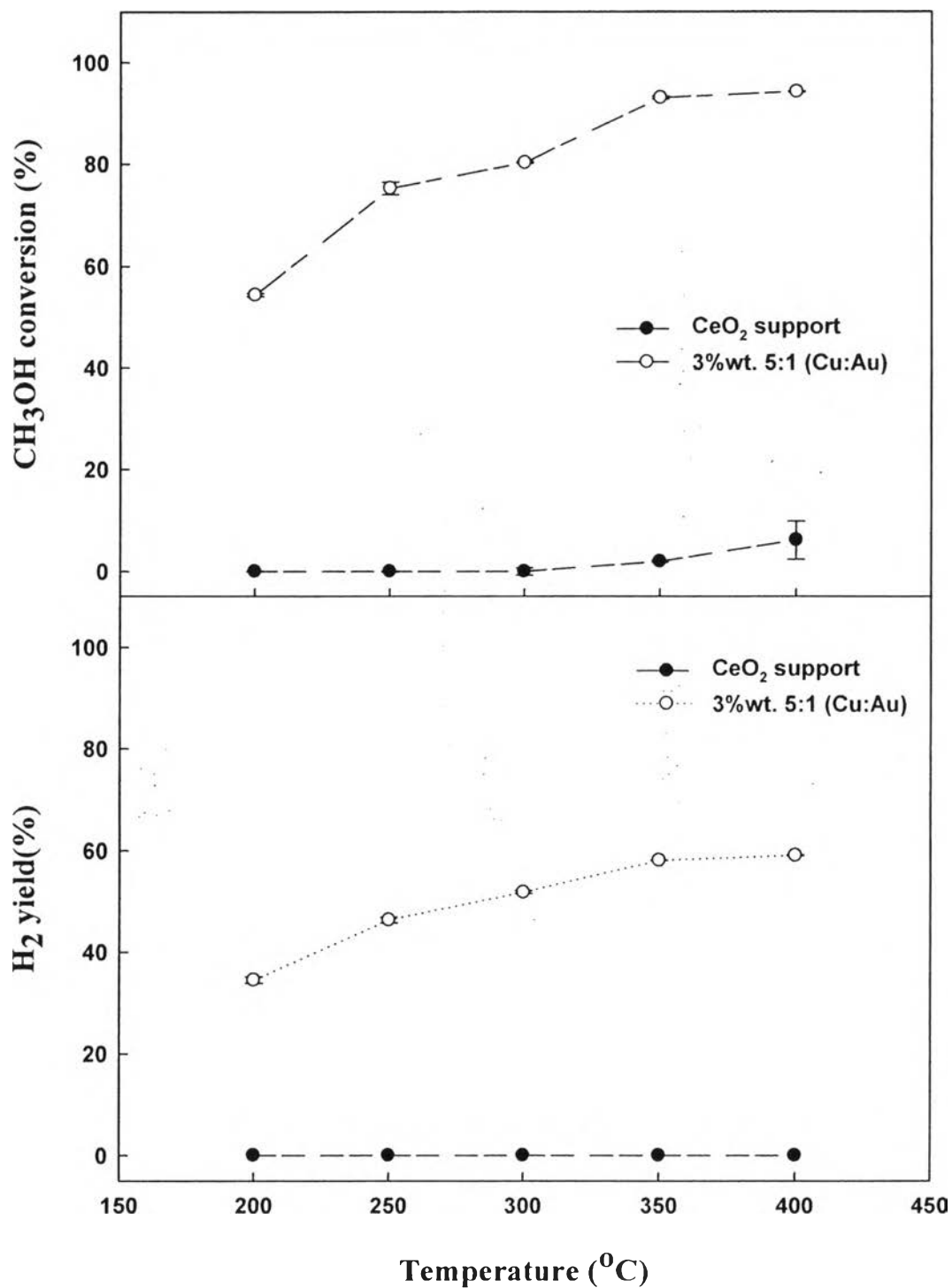


Figure 4.22 Comparison between the catalytic activity of CeO₂ support and 3%wt Au-CuO/CeO₂. Reaction condition: O₂/H₂O/CH₃OH = 1.25/1/1.

4.1.7 Effect of Side Reactions (DM&SRM)

The product of the methanol decomposition is hydrogen which is the main product from the methanol steam reforming. Methanol was decomposed mainly to CO and H₂ over this reaction. Therefore, it is necessary to study the activity of 3%wt 5:1 of Au-CuO/CeO₂ for the methanol decomposition reaction.

Figure 4.24 shows the catalytic activity in the decomposition of methanol reaction, it is clearly seen that the maximum methanol conversion of 54.07% was achieved at 400°C. The methanol conversion was very low, when the reaction was operated at low temperature. It can be concluded that the Au-CuO/CeO₂ was active for both the oxidative steam reforming of methanol reaction and decomposition of methanol reactions.

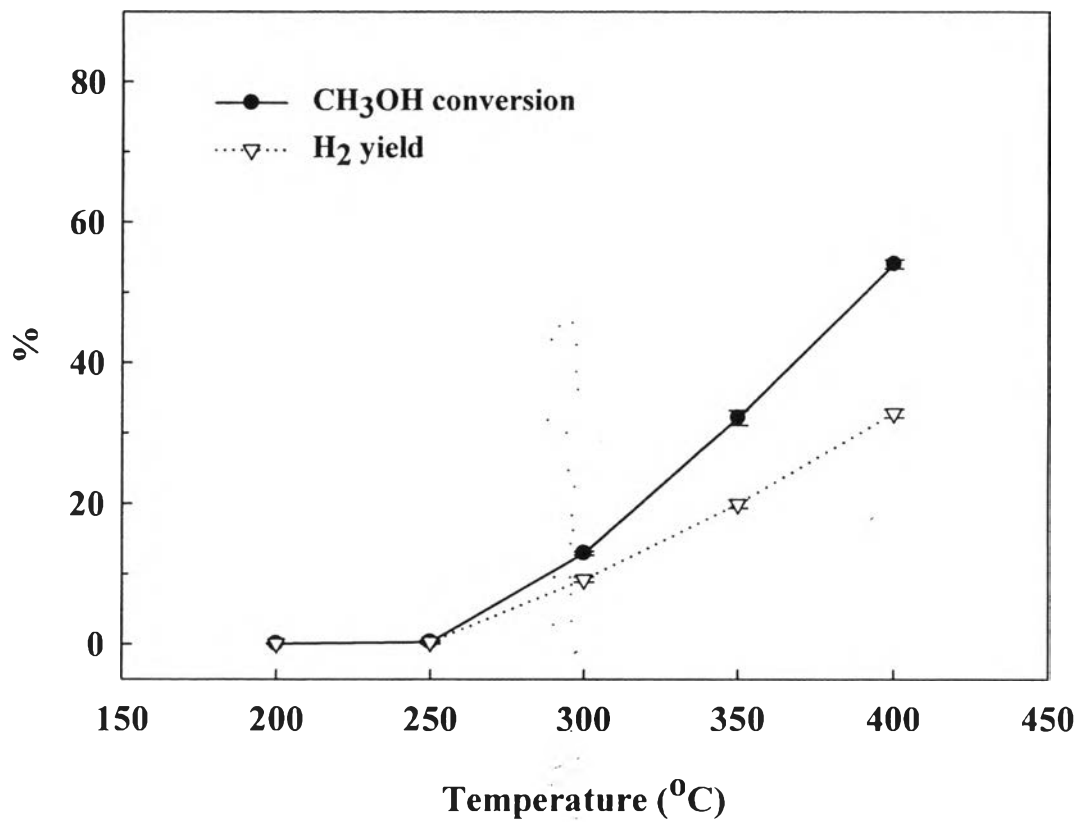


Figure 4.23 Catalytic performance of 3%wt Au-CuO/CeO₂ in the decomposition of methanol reaction.

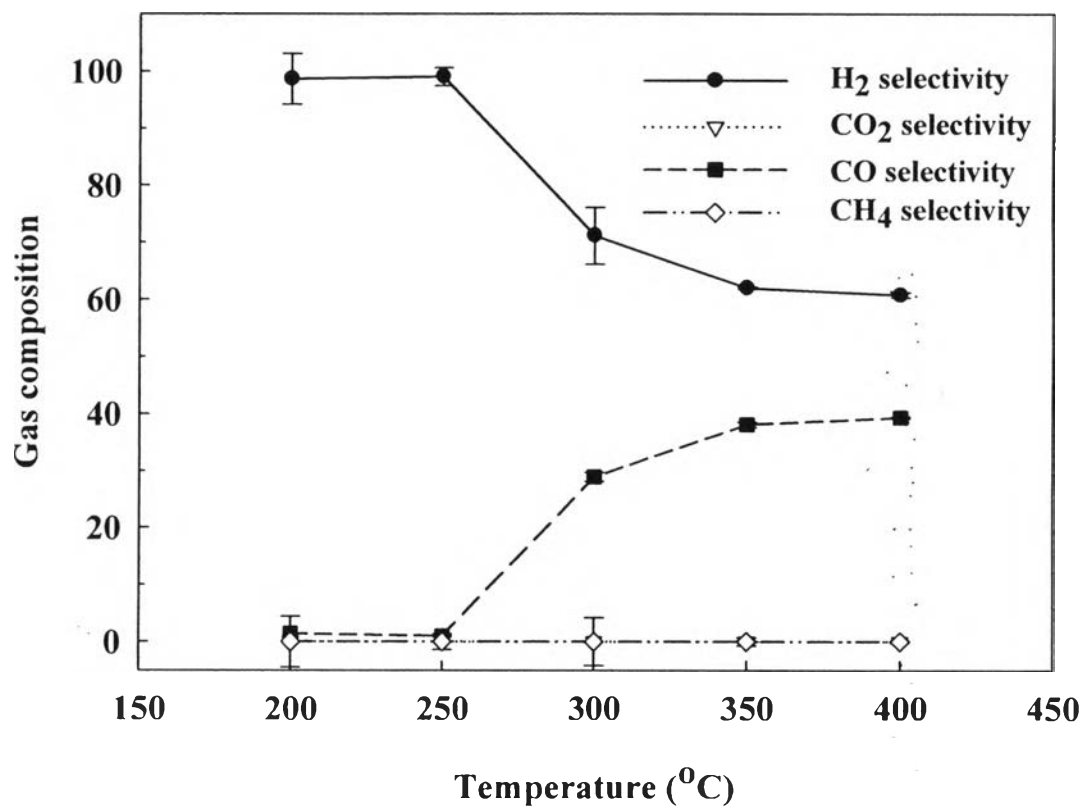


Figure 4.24 Gas composition of 3%wt Au-CuO/CeO₂ in the decomposition of methanol reaction.

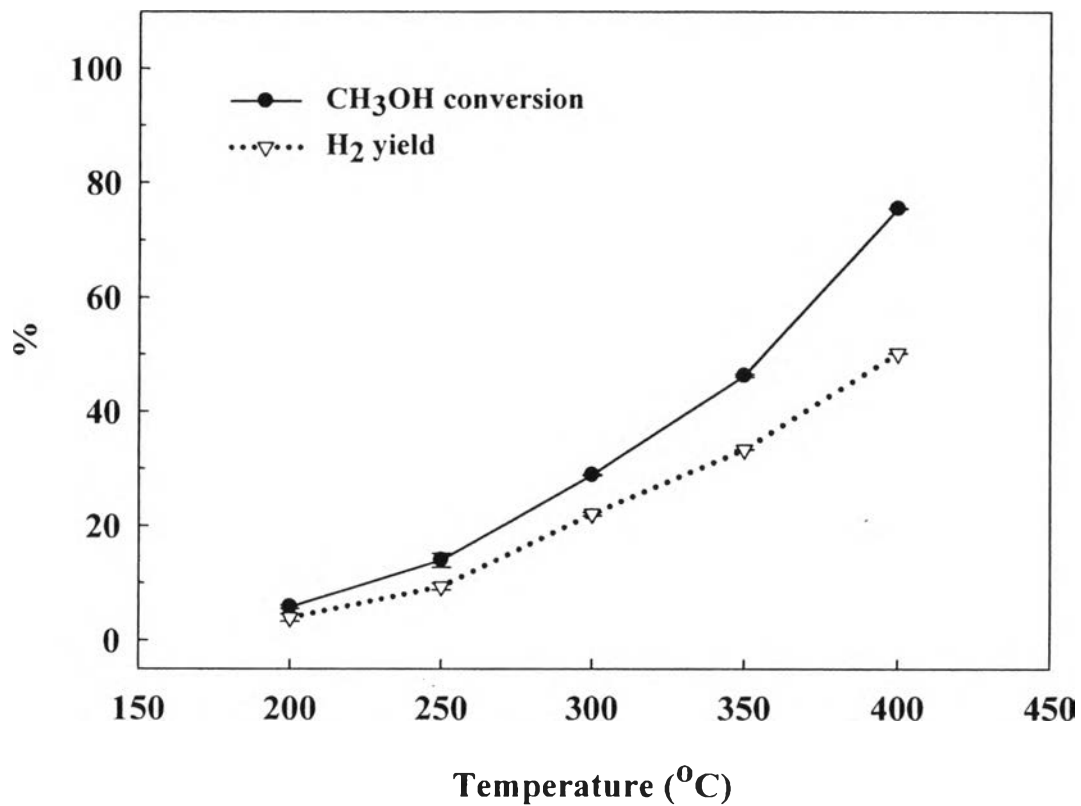


Figure 4.25 Catalytic performance of 3%wt Au-CuO/CeO₂ in steam reforming of methanol reaction.

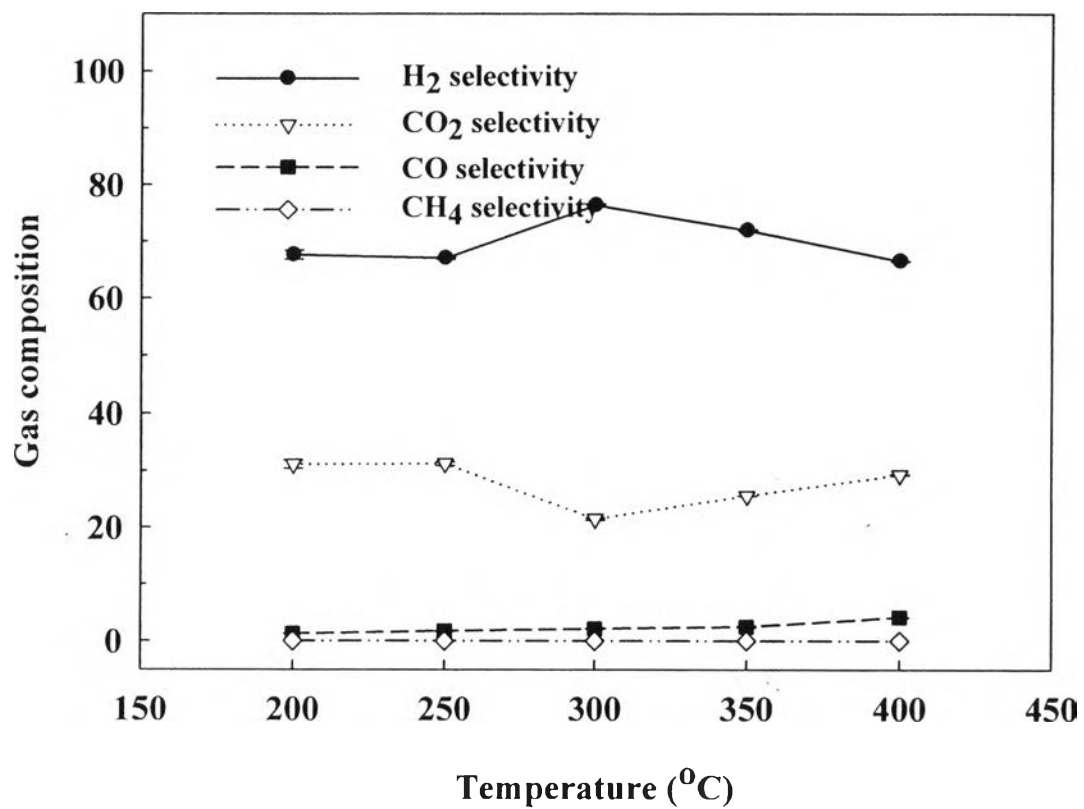


Figure 4.26 Gas composition of 3%wt Au-CuO/CeO₂ in steam reforming of methanol reaction.