CHAPTER IV

RESULTS AND DISCUSSION

The TPD profiles for each adsorbate were obtained for three catalysts, 5.0Pt/Al_2O_3, 1.0Pt-0.1Sn/Al_2O_3, 1.0Pt-0.5Sn/Al_2O_3 (L.C.). In addition, blank experiments were also carried out on Al_2O_3 which had been subjected to the same pretreatment in flowing hydrogen at 673 K as the catalysts.

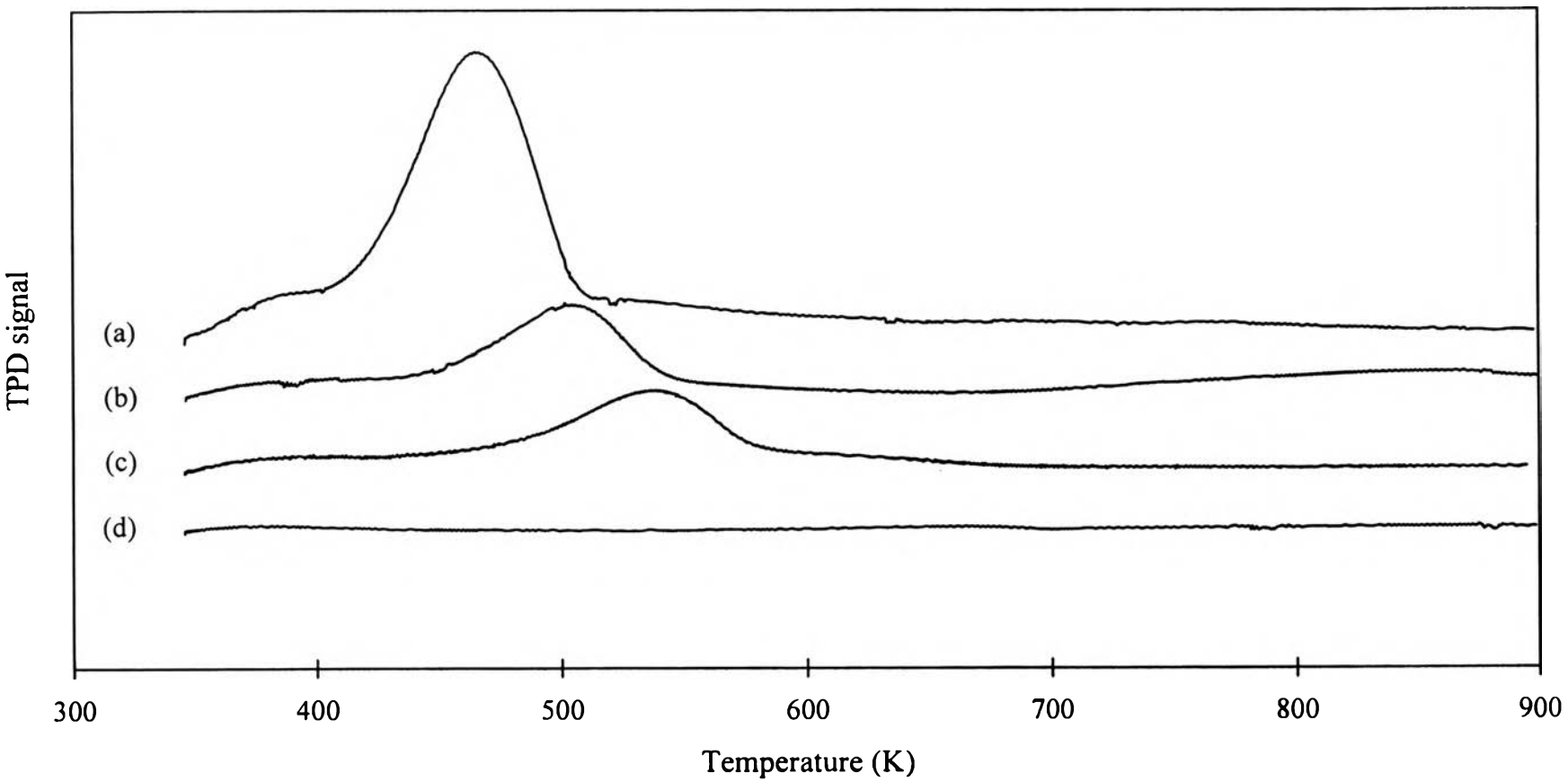
4.1 TPD profile of Methyl Alcohol

Figure 4.1 illustrates the patterns of methyl alcohol desorption on the three catalysts containing different loading of Pt and Sn. After the blank Al_2O_3 exposing to methyl alcohol at 300 K, no desorption peak feature was observed over the entire temperature range up to 900 K, suggesting that the Al_2O_3 support did not adsorb methanol to any significant extent. This is in marked contrast to the observations made by Cordi and Falconer (1996) who found substantial activity of blank alumina for decomposition and dehydration of methyl alcohol. They reported that a small amount of methyl alcohol desorbed over a broad temperature range while most of the methyl alcohol decomposed during TPD between 600 and 900 K, giving primarily CO and H_2, with CO_2 as a minor reaction product. In addition, less than 40 % of the ethyl alcohol dehydrated, giving dimethylether between 500 and 750 K. Cordi and Falconer carried out their experiments on Kaiser A-201 Al_2O_3 which was treated in O_2 at 873 K to dehydrate it before each experiment. The Kaiser Al_2O_3 has a pore volume of 0.46 cm^3/g, an average pore radius of 41 Å, and a surface area of 200 m^2/g (Swecker and Datye, 1990). Our investigation was carried out on nonporous fumed alumina (Degussa) which had a surface area of 90 m^2/g. The alumina consisted of mainly the gamma phase and some of it was in the delta phase. There was less than 0.5 % chlorine present in the alumina. Our

pretreatment was performed in H_2 at 673 K, followed by cooling with nitrogen. Thus, the resulting alumina surface was not dehydrated.

As shown in Figure 4.1, the TPD profiles for all three Pt containing catalyst samples had one major peak. This peak was located at 465 K for $5.0Pt/Al_2O_3$, at 505 K for $1.0Pt\text{-}0.1Sn/Al_2O_3$, and at 540 K for $1.0Pt\text{-}0.5Sn/Al_2O_3$ (L.C.). It is interesting to point out that the $5.0Pt/Al_2O_3$ catalyst had a pronounced shoulder at lower temperatures (around 380-390 K). The other two samples also showed the same manner. The height of the shoulders for all three catalysts were about 1/6 of the height of their main peaks. The $1.0Pt\text{-}0.1Sn/Al_2O_3$ catalyst exhibited an additional broad peak of low intensity starting at around 675 K with a maximum at 860 K. This feature did not appear in the other two catalysts. The TPD peak area of the $5.0\ Pt/Al_2O_3$ catalyst was larger by a factor of 2.19 and 2.74 than that of the $1.0Pt\text{-}0.1Sn/Al_2O_3$ and $1.0Pt\text{-}0.5Sn/Al_2O_3$ (L.C.) samples, respectively. This is in good qualitative agreement with the Pt loading of the samples.

The peak maximum temperature increased going from $5.0Pt/Al_2O_3$ to $1.0Pt\text{-}0.1Sn/Al_2O_3$ and $1.0Pt\text{-}0.5Sn/Al_2O_3$ (L.C.), respectively. This indicates that the bond strength between the adsorbed species and the Pt sites is the weakest for $5.0Pt/Al_2O_3$ and the bond strength increases significantly with addition of tin.

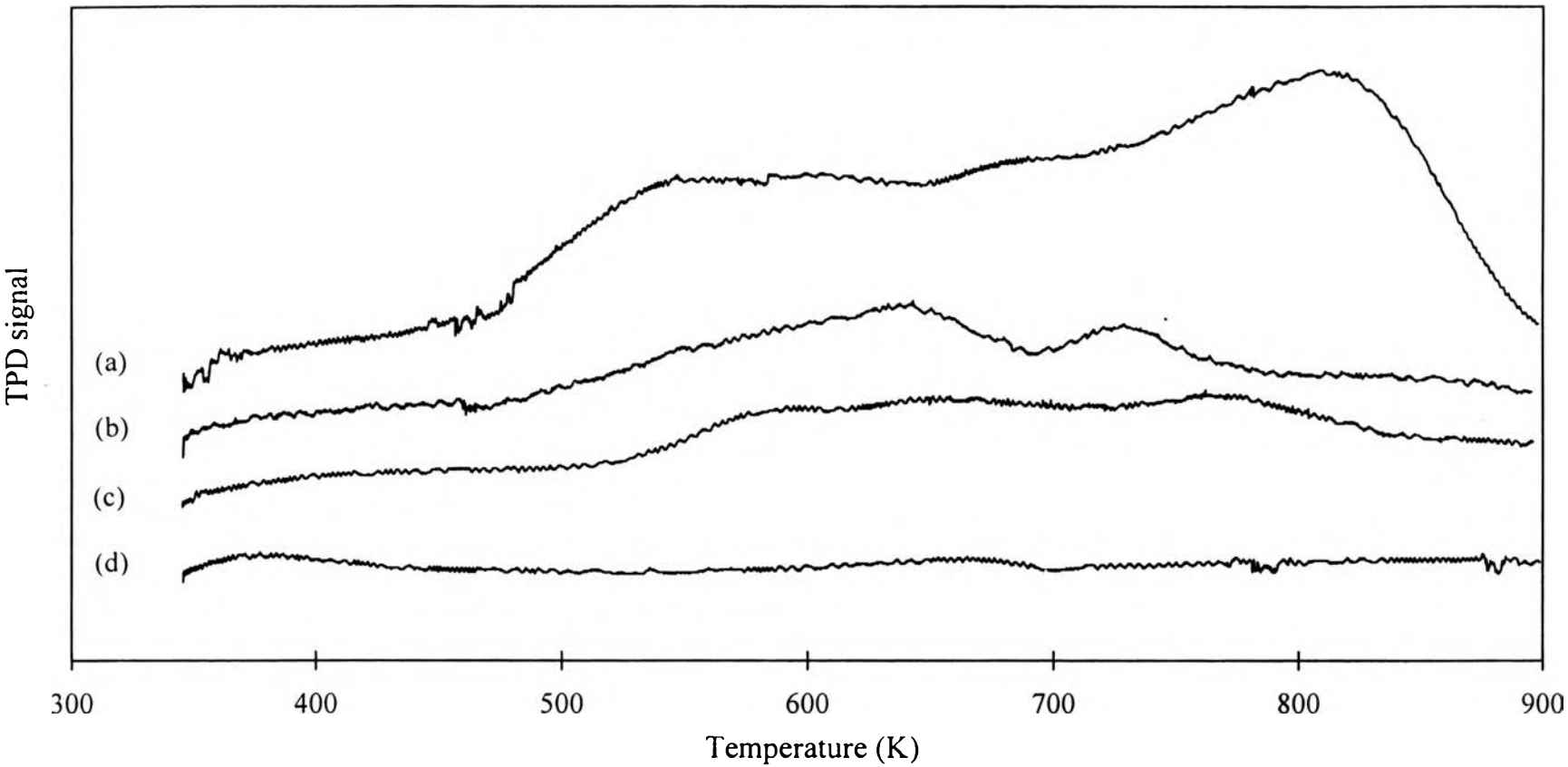


(a) 5.0Pt/Al_2O_3 (b) 1.0Pt-0.1Sn/Al_2O_3 (c) 1.0Pt-0.5Sn/Al_2O_3 (L.C.) (d) Al_2O_3

Figure 4.1 Temperature-Programmed Desorption of methyl alcohol on alumina surfaces containing different loadings of Pt and Sn.

4.2 TPD profile of Acetone

Figure 4.2 shows that the TPD profile of the 5.0Pt/Al_2O_3 catalyst exhibited a broad desorption peak which can be deconvoluted into three sections, with maxima at 545 K, 695 K, and 805 K. The peak at 805 K had the highest intensity. The TPD profile of the 1.0Pt-0.1Sn/Al_2O_3 catalyst also exhibited again a broad peak; starting at about 450 K and reaching a first maximum at about 650 K, followed by a second peak at 730 K. From the spectrum, it is difficult to tell whether or not the broad TPD feature contains a third TPD peak at 550 K. The TPD profile of the 1.0Pt-0.5Sn/Al_2O_3 (L.C.) catalyst had a broad feature with weak peaks ranging from 585 K to 765 K. It can be said that the TPD profile of the 1.0Pt-0.5Sn/Al_2O_3 (L.C.) catalyst had three peaks at 585 K, 650 K, and 765 K. The first peak of the 5.0Pt/Al_2O_3 catalyst reached its maximum at 545 K which was lower than the temperatures of the first peak maximum of the 1.0Pt-0.1Sn/Al_2O_3 and 1.0Pt-0.5Sn/Al_2O_3 (L.C.) but the second and third peaks of the 5.0Pt/Al_2O_3 catalyst are located at higher temperatures than the corresponding peaks of the other two catalysts. If one assumes that the broad TPD feature obtained from the 1.0Pt-0.5Sn/Al_2O_3 (L.C.) catalyst should have three peaks similar to the TPD profile of the 5.0Pt/Al_2O_3 catalyst. Therefore, the second and third peaks should be located at temperatures very similar to those observed on the 5.0Pt/Al_2O_3 sample. However, when the 1.0Pt-0.1Sn/Al_2O_3 is compared to the 1.0Pt-0.5Sn/Al_2O_3 (L.C.), the peak temperature appears to increase with addition of tin. Again, in the case of acetone, there were no significant desorption profiles observed for the blank Al_2O_3.



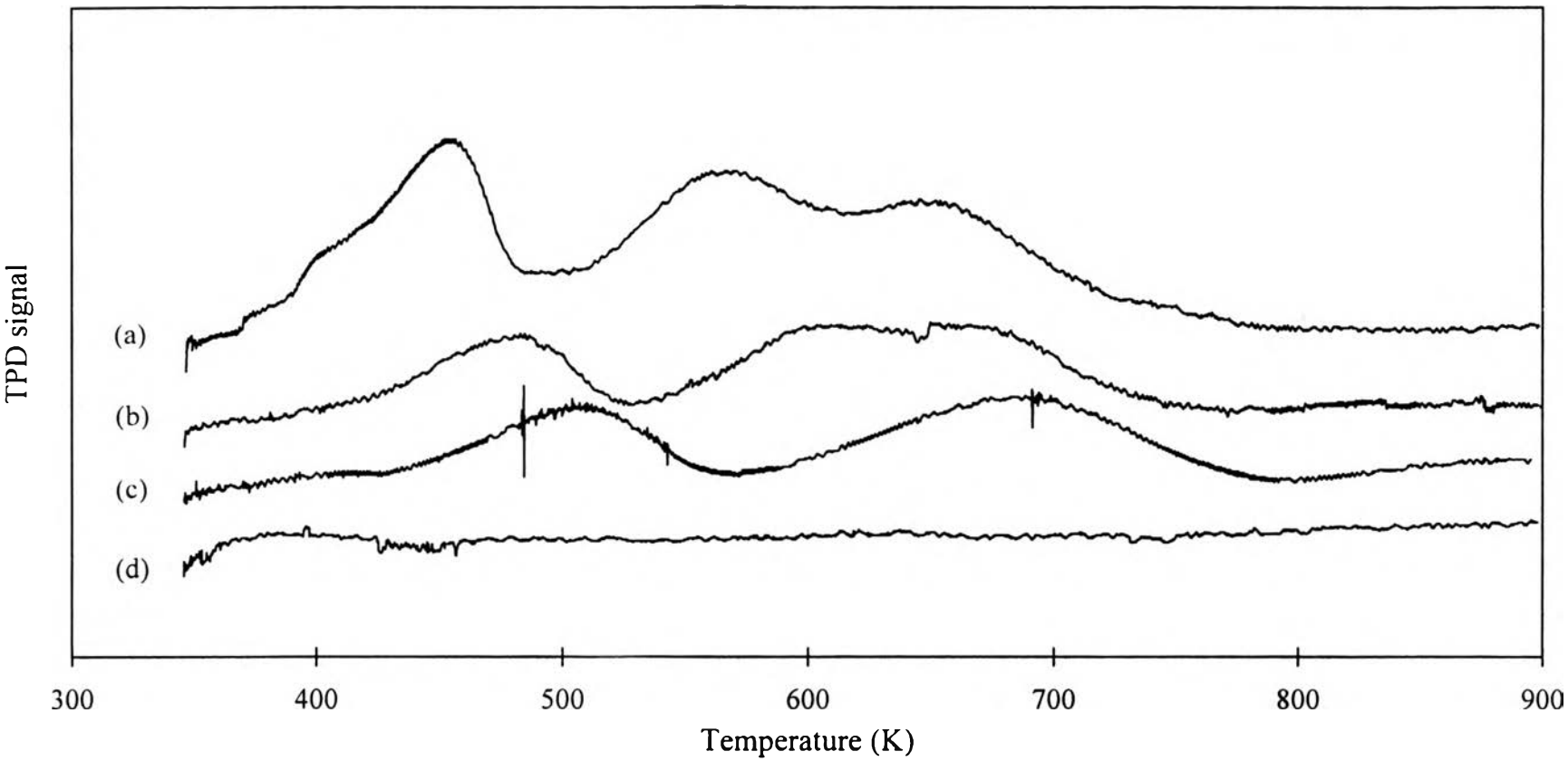
(a) 5.0Pt/Al_2O_3 (b) 1.0Pt-0.1Sn/Al_2O_3 (c) 1.0Pt-0.5Sn/Al_2O_3 (L.C.) (d) Al_2O_3

Figure 4.2 Temperature-Programmed Desorption of acetone on alumina surfaces containing different loadings of Pt and Sn.

4.3 TPD profile of Ethyl Acetate

Figure 4.3 shows that the TPD profile for the 5.0Pt/Al_2O_3 catalyst has three clearly resolved peaks at 450 K, 560 K, and 645 K while the other two catalysts (1.0Pt-0.1Sn/Al_2O_3 and 1.0Pt-0.5Sn/Al_2O_3 (L.C.)) have broad TPD profiles. For the 5.0Pt/Al_2O_3 catalyst, the first peak at the lowest temperature exhibited a distinct shoulder. The TPD profile of the 1.0Pt-0.1Sn/Al_2O_3 catalyst had two peaks at 480 K and 590-670 K. The second peak is very wide. The TPD profile of the 1.0Pt-0.5Sn/Al_2O_3 (L.C.) catalyst exhibited two peaks at 500 K and 680 K. Again the pure alumina pellets did not show a TPD profile. This indicates clearly that there is no adsorption of ethyl acetate on the alumina surface.

From the TPD profiles of the three studied VOCs (methyl alcohol, acetone, and ethyl acetate), it can be pointed out that all the studied VOCs could adsorb on the alumina supporting surface containing Pt. While the pure alumina did not show any adsorption and desorption of the studied VOCs. The overall desorption profiles have shown a trend towards higher temperature following the order of 5.0Pt/Al_2O_3, 1.0Pt-0.1Sn/Al_2O_3, and 1.0Pt-0.5Sn/Al_2O_3 (L.C.), respectively. An addition of tin could increase the temperatures of the desorpion peak.



(a) 5.0Pt/Al_2O_3 (b) 1.0Pt-0.1Sn/Al_2O_3 (c) 1.0Pt-0.5Sn/Al_2O_3 (L.C.) (d) Al_2O_3

Figure 4.3 Temperature-Programmed Desorption of ethyl acetate on alumina surfaces containing different loadings of Pt and Sn.