Properties of Highly Dispersed Cobalt on Zeolites and Manganese on Magnesium Oxide

In the study of highly dispersed cobalt on NaY zeolite, the catalytic performance of the cobalt nanoparticles (Co/NaY) was investigated. The X-ray absorption near edge structure (XANES) was used to characterize the electronic structure of the Co nanoparticles. The results showed that the Co nanoparticles were well-dispersed on the NaY zeolite, with a high degree of dispersion, as evidenced by the absence of any distinct Co peaks in the XANES spectra. The high dispersion of the Co nanoparticles on the NaY zeolite resulted in improved catalytic activity, as evidenced by the increased hydrogenation rates of the model compounds.

Similarly, the catalytic performance of the cobalt on ZSM-5 (Co/ZSM-5) was also assessed. The XANES spectra showed a similar distribution of the Co nanoparticles on the ZSM-5, with a high degree of dispersion. The catalytic activity of the Co/ZSM-5 was also improved compared to the unsupported Co catalyst.

The catalytic performance of the manganese on MgO (Mn/Hi-Mol) was also evaluated. The XANES spectra showed a distinct change in the electronic structure of the Mn nanoparticles on the MgO, indicating a change in the electronic environment. The catalytic activity of the Mn/MgO was also improved, as evidenced by the increased hydrogenation rates of the model compounds.

Overall, the study demonstrated the potential of highly dispersed Co and Mn as catalysts for hydrogenation reactions, with the XANES technique providing valuable insights into the electronic structure and catalytic performance of these systems.
และ Mn(CO)₅CH₃ เมื่อนำมาวางด้วยรับ แล้ววิเคราะห์มีปิโตรซิิสที่อยู่บนผิวหน้า ด้วยเทคนิคสเปกตรโศปิโตรซิิสของการดูดกลืนรังสีอินฟราเรด และอีกสิ่งหนึ่งทราบงานบานเกิดการใช้แนวทางบวกว่าสิ่งที่สับสนที่มีดีต่ำของตัวอย่างที่เตรียมจาก Mn₂(CO)₁₀ และ Mn(CO)₅H ประกอบด้วยสารประกอบแม่เหล็กนิศตร์บันธิมหายชนิดสมกัน การนำบับตัดตัวอย่างต่ำกว่าในภาวะออกซิเจนที่อุณหภูมิต่ำพบว่า เหลือเพียงแม่เหล็กนิศตร์บันธิมหายต่ำภายนอกผิวของ MgO และเซ็กซี่กัลเลชันของแม่เหล็กนิศตร์ที่ด้อยความจุการนำบับตัดตัวออกซิเจนนั้นมีค่ามำกว่าสองในขณะที่ตัวอย่างที่เตรียมจาก Mn(CO)₅CH₃ แต่แม้เกิดนี้อยู่ในตำแหน่งนี้ มีเพียงแม่เหล็กนิศตร์บันธิมหายต่ำภายนอกผิวที่นำบับตัดตัวออกซิเจนที่อุณหภูมิทำไม่ทำให้เกิดการเปลี่ยนแปลงที่น้ำด้านของเซ็กซี่กัลเลชันและจำนวนผลิตภัณฑ์บันธิมหาย

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Zeolite NaY was synthesized by a hydrothermal method and used as a catalyst support. The optimum synthesis condition was 1-day aging time at room temperature and 1-day crystallization time at 100°C. By variation of the synthesis conditions, zeolite NaY transformed to zeolite NaP. The change in the zeolite phase was confirmed by powder X-ray diffraction. The aging time showed a slight effect on the transformation but the crystallization time, with a fixed aging time at 1 day, caused a significant transformation. The NaY phase was not observed after a crystallization time of 4 days and zeolite with NaP as a major phase was obtained after crystallization of 5 days.

The zeolite NaY was used as a support for cobalt catalysts (Co/NaY) in n-butane hydrogenolysis and compared to that supported on zeolite ZSM-5 (Co/ZSM-5). The conversion of n-butane from Co/NaY was lower than that from Co/ZSM-5, consequently, further study was done only over Co/ZSM-5. Because metallic cobalt is an active form in n-butane hydrogenolysis, the reducibility of cobalt catalysts was determined by X-ray absorption near edge structure. In monometallic catalysts (1, 4, 7 and 10 wt% Co/ZSM-5), the reducibility and conversion of n-butane
hydrogenolysis increased with the cobalt content. Addition of 1 wt% Pd to the monometallic cobalt did not significantly enhance the reducibility and catalytic activity. Monometallic cobalt catalysts produced only methane whereas bimetallic ones produced trace amount of ethane and propane with methane as main product.

Another part of this work focused on improvement of uniformity and dispersion of manganese supported on magnesium oxide (MgO) prepared from organometallic complexes as metal precursor by chemical vapor deposition. Mn$_2$(CO)$_{10}$, Mn(CO)$_3$H and Mn(CO)$_3$CH$_3$ compounds were deposited on MgO and the resulting surface species were characterized by X-ray absorption, electron paramagnetic resonance, and infrared spectroscopies. Deposition of Mn$_2$(CO)$_{10}$, Mn(CO)$_3$H on MgO resulted in mixed manganese carbonyl species. Treatment of sample prepared from Mn$_2$(CO)$_{10}$ and Mn(CO)$_3$H on MgO in flowing O$_2$ at room temperature resulted in formation of manganese tricarbonyl species with the oxidation state of manganese higher than +2. In contrast, chemical vapor deposition of Mn(CO)$_3$CH$_3$ on MgO led to the formation of only one surface species, manganese tetracarbonyl, in which the manganese was cationic. The oxidation state and number of carbonyl ligand of manganese remained unchanged after this sample was treated in O$_2$. 

School of Chemistry                      Student’s Signature________________________

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