TOP 10 RESEARCH ACHIEVEMENTS

Heterogeneous Metal Atomic Catalysts for Gas-Phase Pollution Remediation

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Automobile emission contains toxic environmental pollutants, such as CO, hydrocarbon(C_3H_6, C_3H_8) and NO_x, and they should be purified by heterogeneous catalysts. Typical automobile catalysts have showed insufficient performance for pollution remediation due to poor activity at low temperature(≤150 °C) and low durability under hydrothermal aging(≥750 °C, 25 h). In this research, metal(Pt, Pd, Rh) atomic catalysts were fabricated, and they showed superior low-temperature activity. Furthermore, they exhibited excellent durability under hydrothermal aging, long-term reaction and recycling test. Thus, this technology would lead the field of automobile catalysts, and have tremendous academic and industrial values.

1. Background

Carbon monoxide (CO), hydrocarbons (C_xH_y), and nitrogen oxides (NO_x) emitted from automobile exhaust gas are toxic molecules to the human respiratory system, and are representative atmospheric environmental pollutants that become precursors of fine dust and smog. These pollutants should be purified to nitrogen (N_2), water (H_2O), carbon dioxide (CO_2) through oxidation and reduction reactions using heterogeneous metal catalysts. However, conventional catalysts have showed poor activity at low temperatures (below 150°C) and poor durability at high temperatures (above 750°C, with humidity). Specifically, for gasoline hybrid vehicles, it is important to develop a highly active metal catalyst working at low temperatures because the temperature of the exhaust catalyst layer is lowered while the fuel efficiency is greatly enhanced. At the same time, high temperatures are accompanied during fuel combustion process in the engine, so the catalyst structure should have high durability even in high temperature condition.

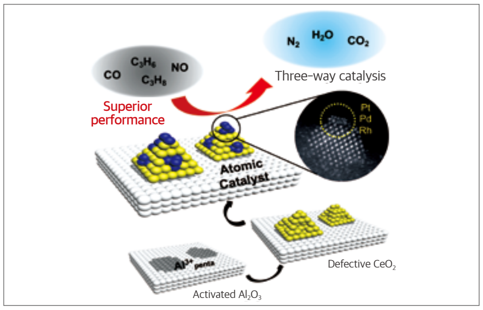
2. Contents

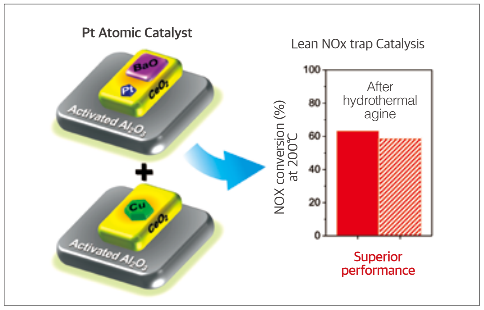
Support design is mostly important to fabricate the metal atomic catalysts, which are all the metal atoms are exposed on the surface (100% metal dispersion) with strong metal-support interaction. The structure of metal atom is determined as the characteristics of the support, and then the activity and durability of the metal catalyst would be changed. In this study, Prof. Hyunjoo Lee research group devised the highly defective reducible metal oxide supports to fully disperse and strongly anchor the metal atoms. The reducible metal oxide has a characteristic that the oxidation number of the metal is easily changed depending on the conditions, resulting in oxygen vacancy defect sites. In this study, a large amount of coordinatively unsaturated Al^{3+}_{penta} species, which are strong anchoring sites, were created by pre-reducing the alumina (γ-Al_2O_3) support. Then, ceria (CeO_2) was impregnated to the Al^{3+}_{penta} sites, resulting in alumina-ceria dual structure support. Strong interaction of alumina-ceria produced the highly defective ceria with many Ce^{3+} sites. When the metals (Pt, Pd, Rh) were impregnated to the defective ceria, exceptionally strong interaction of metal-ceria effectively lowered the surface free energy of labile metal atoms. As a result, metal atomic catalysts (Pt, Pd, Rh/CeO_2-Al_2O_3) were designed with 100% metal atom dispersion.

Metal atomic catalysts were employed for gas-phase pollution remediation of three-way catalysis (TWC) and lean NO_x trap catalysis (LNT). TWC is a simultaneous surface reaction of oxidation of CO and C_xH_y, and reduction of NO_x. Metal atomic catalysts exhibited the world's best low-temperature activity, and showed higher activity and durability than commercial TWC catalyst. Specifically, all of CO, C_3H_6, and NO were completely removed below 150℃, and conversion of C_3H_8, which is difficult to remove, reached to 100% below 300℃. Furthermore, metal atomic catalysts showed excellent durability against hydrothermal aging (900℃, 24h), long-term reaction (150℃, 420h), recycle test without any degradation. In LNT, NO_x is trapped on the catalyst surface under fuel-lean condition, and then NO_x is catalytically reduced to N_2 under fuel-rich condition. Metal atomic catalysts showed 3 times higher performance than commercial LNT catalyst for low-temperature NO_x removal below 200℃. In addition, even after hydrothermal aging (750℃, 25h), the structure of the metal atomic catalyst was unchanged, and 93% of the initial performance was achieved.

3. Expected effects

Prof. Hyunjoo Lee research group developed the novel concept of metal atomic catalysts with superior performance for gas-phase pollution remediation. Metal atomic catalysts, which have 100% dispersion by strong interaction with defect sites of defective support, showed the world's best activity and durability. Thus, this technology would lead the future in the automobile catalyst field, and present guidelines for the development of highly active and durable heterogeneous metal catalysts. This technology can be applied for modern gasoline hybrid vehicles as it can efficiently deal with continuously strengthened environmental regulations. Furthermore, it would be possible to apply this technology to all industries requiring exhaust gas purification such as ship, aviation, motorcycles, and power plants. The metal atomic catalysts have infinite academic and industrial values in heterogeneous catalysis, and would make great impacts on the future society.





Research outcomes

[Paper] H. Jeong, O. Kwon, B.-S. Kim, J. Bae, S. Shin, H.-E. Kim, J. Kim, H. Lee*, "Highly Durable Metal Ensemble Catalysts with Full Dispersion for Automotive Applications beyond Single-Atom Catalysts", Nature Catalysis 3, 368-375 (2020) [2020 Impact Factor = 30.471]

B.-S. Kim, H. Jeong, J. Bae, P. S. Kim, C. H. Kim, H. Lee*, "Lean NO_x Trap Catalysts with High Low-Temperature Activity and Hydrothermal Stability", Applied Catalysis B: Environmental 270, 118871 (2020) [2020 Impact Factor = 16.683]

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