Support-shape Dependent Catalytic Activity in Pt/alumina Systems Using USANS/SANS

Sang Hoon Kim[a*], Sugyeong Han[b], Heonphil Ha[a], Jiyoung Byun[a], Man-ho Kim[b]
[a]*Center for Materials Architecturing, KIST, Seoul, 136-791 Korea*
[b]*Advanced Analysis Center, KIST, Seoul, 136-791 Korea*
[*]*Corresponding author: kim_sh@kist.re.kr*

1. Introduction

Pt nanoparticles dispersed on ceramic powder such as alumina and ceria powder are used as catalyst materials to reduce pollution from automobile exhaust, power plant exhaust, etc. Much effort has been put to investigate the relationship between types of catalyst support materials and reactivity of the supported metallic particles. In contrast to this, studies on the relationship between shapes of the support materials and reactivity of the supported metallic particles are rare. Since Van der Waals forces between fine particles are affected by the shape, the dispersion of the particles can be dominated by the shapes. The surface shape of support materials can also be expected to control the catalysts size with the surface shape of support materials.

In this presentation, we show our SANS (small angle neutron scattering) –USANS (ultra small angle neutron scattering) analysis on the structural differences of different shapes of the same γ alumina powder with different loadings of Pt nanoparticles. Then, the reactivity of the prepared catalyst materials are presented and discussed based on the investigation of the structure of the support materials by SANS[1].

2. Methods and Results

2.1 Neutron scattering analysis

The Microscopic structures of the two types of γ-alumina, AA and AO, were characterized with USANS and SANS measurements. Independent measurements of the USANS and SANS superimpose in the double logarithm plot of the total cross-section against Q (Figure 1 for AA and Figure 2 for AO). The USANS-SANS scattering profiles cover more than 11 orders in magnitude of total scattering cross-sections (y-axis) and 4 orders in Q scale, which range from nanometers to micrometers in size.

2.2 Catalytic reactivity comparisonl

To find a correlation between the support shape and deposition characteristics of Pt catalyst nanoparticles, we deposited Pt on AA and AO by arc plasma deposition (APD) and checked the deposition behavior. Actual Pt loading amounts measured by ICP-MS were 2.01 wt% for AO and 2.31 wt% for AA.

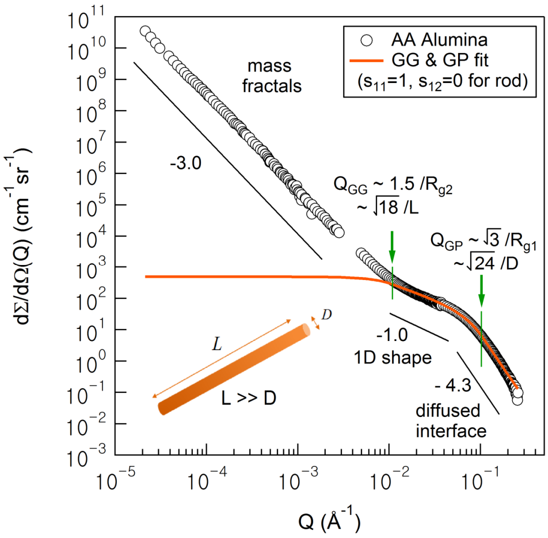

Fig. 1. USANS and SANS profiles (open circle) of pure AA alumina and generalized Guinier-Power and Guinier-Guinier fit (line) with s_{11}=1, s_{12}=0. Inset is a rod shape.

After Pt deposition, we investigate catalytic activity of the Pt/alumina systems with two different γ-alumina powders, rod-like and platelet-like. The catalyst samples underwent a model reaction closely related to the three way catalyst (TWC) reactions (CO, NO, C_3H_6 conversions) in a lean condition in a flow reactor [10]. Figure 3 shows CO, NOx, and C_3H_6 conversion rates of the two samples as a function of temperature.

Overall, AA samples showed higher conversion rates than those of AO samples. This observation is in line with the observation that the dispersion of Pt nanoparticles on AA was better than that on AO (not shown), leading to higher active surface area on AA. Especially, low temperature efficiency of the AA sample was particularly higher than those of the AO samples. In Figure 3(a), catalytic efficiency of Pt-AA for CO oxidation was about 62%, while that of Pt-AO was about 23 % at 250 C. In other words, Pt-AA had more than 100% higher catalytic efficiency than Pt-AO at 250 C. This superior catalytic efficiency was observed at 200 - 250 C. Other reactions such as NO reduction (Figure 3(b)) and C_3H_6 oxidation (Figure 3(c)) showed similar tendencies. Given the same materials of the catalyst (Pt) and support (γ-alumina), the exposed surface area of the deposited Pt nanoparticles will be one of the most important factors for catalytic activity.

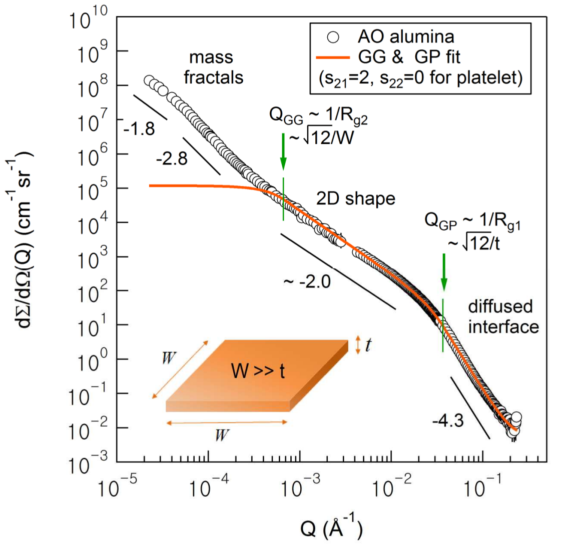

Fig. 2. USANS and SANS profiles (open circle) of pure AO alumina and generalized Guinier-Power and Guinier-Guinier fit (line) with s_{21}=2, s_{22}=0. Inset is a simplified platelet with the same lateral dimension.

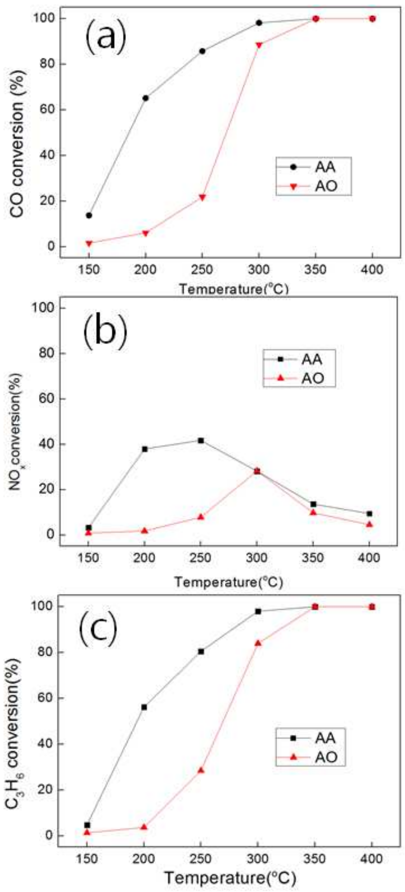

Fig. 3. Conversion rate comparisons for TWC model reactions. (a) CO oxidation, (b) NO reduction, (c) C_3H_6 oxidation.

3. Conclusions

The shapes of gamma alumina, rod-like or plate-like shape, were determined from nanometer to micrometer with USANS and SANS analysis. We found that the platelet-like alumina consists of an aggregate of 2 ~ 3 layers, which further reduce specific surface area and catalytic activity compared to rod-like shape. Rod-like shape shows more than 100% enhancement in the catalytic activities in model three-way-catalyst (TWC) reactions of CO, NO, and C3H6 at low temperature around 200 ℃.

REFERENCES

[1] Sang Hoon Kim, Sugyeong Han, Heonphil Ha, Jiyoung Byun, Man-ho Kim, Support-shape dependent catalytic activity in Pt/alumina systems using ultra-small (USANS) and small angle neutron scattering (SANS), Catalysis Today (accepted).