Direct decomposition of NO over Au-dispersed NiO/$(Y_{0.99}Ba_{0.01})_2O_3$ catalyst

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Abstract: Effects of Au dispersion on direct decomposition of nitric oxide (NO) over $(Y_{1-x}Ba_x)_2O_3$ were investigated and it was found that 1mol% Au-dispersed NiO/$(Y_{0.99}Ba_{0.01})_2O_3$ catalyst shows higher NO decomposition activity at all temperature range examined. The diffraction peaks of Au-$(Y_{0.99}Ba_{0.01})_2O_3$ of XRD patterns were shifted to a slightly lower angle compared with that of $(Y_{0.99}Ba_{0.01})_2O_3$. This shift could be explained by the residual tensile strain, which results from the difference in the thermal expansion coefficients between metal and metal oxides. In this study, effect of tensile strain on NO decomposition activity was investigated.

Keywords: NO decomposition, Au dispersed, $(Y_{1-x}Ba_x)_2O_3$ catalyst.

1. Introduction (11-point boldface)

Nitrogen oxides (NO_X), which are mainly formed by internal combustion engines such as diesel engine, are extremely toxic to a human body and also harmful to environment as a main source of both acid rain and photochemical smog. Among all the developed solutions of NO removal until now, direct decomposition of NO into N_2 and O_2 is considered to be the most ideal way because a reductant is not necessary and the products are harmless [1, 2]. However, for all the catalysts developed for directly NO decomposition up to now, the reaction temperature is still much higher than the practical process. The operation at low temperature is desired because it can lower costs of the total systems. Introducing noble metals have been reported as an efficient way to lower the reaction temperature of the system.

It is well-known that the thermal expansion coefficients of metal and metal oxides are very different. Therefore, when a metal and a metal oxide are connected at an elevated temperature and then cool it down, a large strain will be introduced at the interface between the metal and the metal oxide. According to the difference in thermal expansion coefficients between the metal and the metal oxide, we can expect three-dimensional tensile or compressive strain can be introduced [3]. Therefore, the effect of strain on catalytic activity of NO decomposition is expected because strained lattice increase oxygen diffusion resulting in desorption of oxygen. In this study, we investigated the effect of metallic Au-dispersed $Y(Ba)_2O_3$ catalyst in direct decomposition of NO for improved catalytic performance.

2. Experimental

Au-dispersed $(Y_{1-x}Ba_x)_2O_3$ solids were prepared via solid state reaction [4]. Appropriate amounts of $Y(NO_3)_3{\cdot}6H_2O$ (Kishida, 99.9 wt%), $Ba(CH_3COO)_2$ (Kishida, 99 wt%) and 1mol% $HAuCl_4{\cdot}4H_2O$ (Kishida, 99.0%) were dissolved in distilled water. The solution was heated under stirring until the water evaporated. The gel thus obtained was then subjected to pre-calcination by ventilated oven at 400 °C for 2 h to remove NO_x followed by calcination at 800 °C for 6 h. Then the obtained powder was pressed into a disk 20 mm in diameter and 1 mm thick, and finally the prepared disks were sintered at 1300 °C for 6 h. Au-dispersed NiO/$(Y_{1-x}Ba_x)_2O_3$ samples were prepared by grinding Au-dispersed $(Y_{1-x}Ba_x)_2O_3$ disks into powder and make a solution with proper amount of $Ni(CH_3COO)_2{\cdot}4H_2O$ (Wako, 98 wt%). The solution then evaporated to dryness with heating under stirring and finally calcined at 900 °C for 6 h. Also the samples ($(Y_{1-x}Ba_x)_2O_3$ and NiO/$(Y_{1-x}Ba_x)_2O_3$) without Au dispersed were prepared under the same conditions except

not adding $HAuCl_4 \cdot 4H_2O$. NO direct decomposition was performed by using atmospheric pressure fixed bed reactor and 0.5% NO diluted with He was fed as reactant.

3. Results and discussion

Metallic Au peaks can be observed in the XRD patterns of 1mol%Au-$Y_{0.99}Ba_{0.01}O_{1.5}$ with different sintering temperatures suggesting that Au was dispersed in the prepared samples as metallic state instead of partial substitution of Y_2O_3 lattice. Figure 1 shows the comparison of magnified XRD patterns of the main peak of Y2O3 with and without Au-dispersed catalysts at different sintering temperatures. The peaks belongs to $Y_{0.99}Ba_{0.01}O_{1.5}$ after 1100 °C sintering shifted to low angle with adding 1mol% Au indicating that Y_2O_3 lattice was tensile. However, with increasing sintering temperature, $Y_{0.99}Ba_{0.01}O_{1.5}$ peaks shifted to high angle suggesting that tensile strain formed in the catalyst. Figure 2 shows the comparison of the NO conversion of Au dispersed NiO/$(Y_{0.99}Ba_{0.01})_2O_3$ and NiO/$(Y_{0.99}Ba_{0.01})_2O_3$ catalysts. Both catalysts shows NO decomposition activity at temperature higher than 600 °C, and the oxygen formation was also observed from 600 °C. The catalyst with Au started to show higher nitrogen and oxygen yield than that on non-Au dispersed catalyst from 600 °C and at 850 °C the nitrogen and oxygen yield of Au catalyst was almost twice higher than that of the control catalyst. TEM observation suggests Au exists at the bulk of Y_2O_3 grain and it seems not that the increased NO decomposition acivity can be assigned to catalytic effects of Au. Thus it is considered that the tensile strain introduced at the interface of Au and Y_2O_3 works positively to oxygen desorption resulting in the increased NO decomposition activity of Y_2O_3 added with BaO.

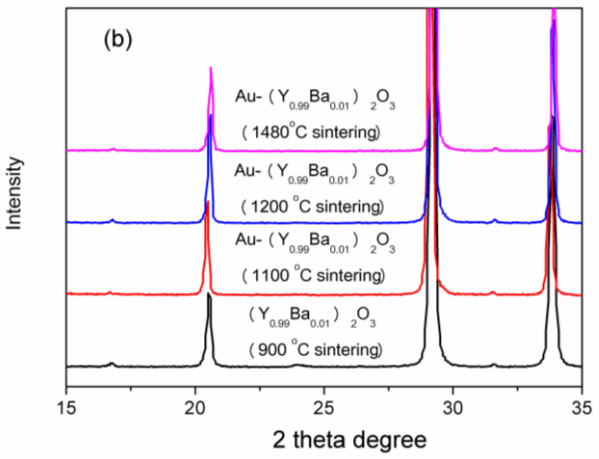

Figure 1. XRD patterns of with and without Au-dispersed samples at different sintering temperatures

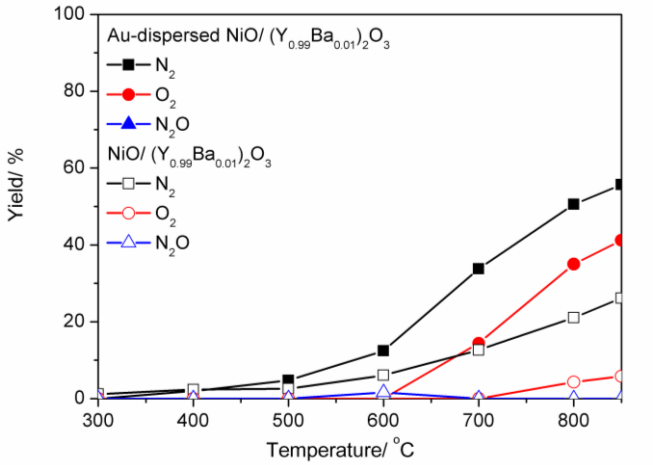

Figure 2. Temperature dependence of N_2 and O_2 yield on Au-NiO/$(Y_{0.99}Ba_{0.01})_2O_3$ and NiO/$(Y_{0.99}Ba_{0.01})_2O_3$ catalysts (P_{NO} = 0.5%, W/F=3.0gscm^{-3}).)

4. Conclusions

Effects of Au dispersed on direct decomposition of NO over $(Y_{1-x}Ba_x)_2O_3$ were investigated and it was found that 1mol% Au-dispersed NiO/$(Y_{0.99}Ba_{0.01})_2O_3$ catalyst shows higher NO decomposition activity, in particular, at 600 °C. The XRD patterns of Au dispersed samples sintering at 1100 °C shift lightly to lower angle showing the introduced tensile strain at the interface of Au particle and $(Y_{0.99}Ba_{0.01})_2O_3$ particle. Therefore, introduction of tensile strain is effective for increasing NO decomposition activity on the Au dispersed catalyst.

References

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