

Effect of hydrothermal aging on Pd/SSZ-13 in low temperature NO adsorption for cold start application

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As the regulation on NO_x emission in exhaust gas has been globally issued and enforced, the purification of NO_x from diesel engine is regarded as a great challenge. Although selective catalytic reduction (SCR) catalyst effectively reduce NO_x in exhaust gas, this system cannot operate properly below 200 °C (i.e. during cold start period, initial ca. 100 – 200 sec of engine operation). Therefore, during the warm-up of NO_x reduction catalyst, the considerable amount of NO_x is emitted into atmosphere without any treatment [1]. Recently, cold start NO_x trap catalysts attract significant attention with a concept of storing NO_x at low temperature (i.e. during cold start) and releasing them at elevated temperature, where SCR operates. In this work, we tried to apply various Pd/SSZ-13 catalysts with different Pd loading method and investigate the effects of hydrothermal aging (HTA) treatment on the NO_x adsorption ability and the physicochemical change of Pd/SSZ-13.

Figure 1a shows the NO_x desorption curves of fresh and HTA Pd(2)/SSZ-13 IWI catalysts (prepared by Incipient Wetness Impregnation) after NO adsorption for 100 sec. It must be pointed out that the NO adsorption ability of fresh Pd(2)/SSZ-13 catalyst is negligible, as evidenced by NO_x desorption curve in the Figure 1a that exhibits only small NO_x desorption peak at around 350 °C. In contrast to fresh catalyst, HTA Pd(2)/SSZ-13 IWI catalyst clearly exhibits two distinct desorption peaks having a maximum intensity at around 250 °C and 400 °C, indicating that HTA treatment allows catalyst to have the considerable NO adsorption ability at low temperature. Regardless of Pd loading method, WET, ION, and S-S catalysts (prepared by wet

impregnation, ion exchange, solid-state ion exchange, respectively) exhibit the similar NO adsorption/desorption behavior with IWI sample. Combined H₂-TPR and Pd EXAFS results clearly demonstrate that the main state of Pd in fresh catalysts is PdO which cannot adsorb NO [2], explaining the reason why fresh catalyst has the insignificant NO adsorption ability, whereas HTA treatment on Pd/SSZ-13 induces the redistribution of PdO to produce Pd ion species in SSZ-13 zeolite.

Figure 1b shows DRIFT spectra of HTA Pd(2)/SSZ-13 IWI catalysts acquired during NO_x desorption after NO adsorption at 120 °C for 1 hr. NO adsorbed HTA catalyst exhibits two ionic Pd-NO complex peaks at 1810 and 1860 cm⁻¹, demonstrating the presence of Pd ion species in HTA Pd/SSZ-13. In Figure 1b, during the temperature ramping from 120 to 300 °C, the peak at 1810 cm⁻¹ significantly decreases, indicating that the low temperature NO_x desorption peak (at 250 °C) arises from the ionic Pd-NO species at 1810 cm⁻¹. Above 300 °C, since nitrosyl complex at 1860 cm⁻¹ begin to decrease, the desorption of NO between 300 and 500 °C is mainly related to the band at 1860 cm⁻¹. Combined NO adsorption/desorption and DRIFT results clearly indicate that ionic Pd species are the active site for NO_x storage. In summary, HTA treatment on Pd/SSZ-13 transform PdO into Pd ion species which play as the active site for low temperature NO adsorption.

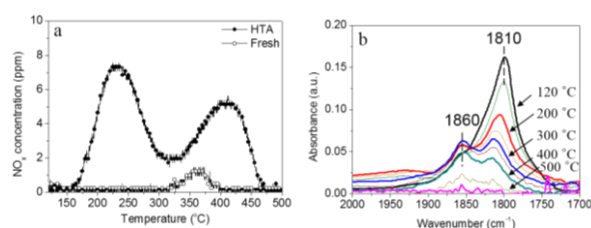


Figure 1 (a) NO_x-TPD curves of fresh and HTA Pd(2)/SSZ-13 IWI and (b) DRIFT spectra of HTA Pd(2)/SSZ-13 IWI catalyst during NO_x-TPD after NO adsorption for 1 hr.

REFERENCES

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