Controlled Synthesis of Nanoscale Pd-CeO_2 Structures and Their Corresponding Influence on Catalytic Performance

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Abstract: We applied two different methods to obtain nanoscale Pd-ceria structures. In one sample core-shell like Pd-CeO_2 was obtained to maximize the Pd-ceria interface. In other sample, CeO_2 nanocrystals merely decorated on Pd NPs. Through CO oxidation and CH_4 combustion, we demonstrated that the Pd-CeO_2 interface contributed to the low-temperature CO oxidation process while had detrimental effect on low-temperature CH_4 combustion. For CH_4 combustion we believed PdO was the active component, minor CeO_2 decoration had little influence on the CH_4 combustion activity. The experiment results deepen the understanding of the nature of active sites and provide guidance on designing novel nanocatalysts.

Keywords: Pd-CeO_2, nanostructure, CH_4 combustion, CO oxidation

1. Introduction (11-point boldface)

In the past decade, the concept of 'nanocatalyst', with the emphasis on the controlled fabrication of active sites at nanoscale level, has emerged and developed rapidly[1]. In this Abstract, we introduced new methods to obtain different 'dispersible' Pd-ceria nanostructures by using nanoparticles as 'artificial atoms'. The catalytic performances in CO oxidation and methane combustion reactions were evaluated and the influence of specific surface nanostructure was observed.

2. Results and discussion

Two model samples (denoted as MS-1 and MS-2) were prepared. In MS-1 we used 2.5nm Pd nanoparticles as starting building block and the CeO_2 nanoparticles were functionalized via an amino-assisted approach at room temperature, forming a core-shell like structure (Fig 1a). In MS-2 4.5nm Pd nanoparticles were used, and CeO_2 nanocrystals were introduced by thermal decomposition method (Fig 1c).

Both sample were supported on Al_2O_3 and calcined at 550 °C. In MS-1, small Pd nanoparticles were mostly encapsulated in CeO_2 nanoparticles while in MS-2 CeO_2 nanocrystals were scattered randomly, with PdO nanoparticles were the dominate species (Figure1b&d). In both samples the size of the Pd NPs could be well maintained and the schematic illustration of MS-1&MS-2 was given in Scheme 1.

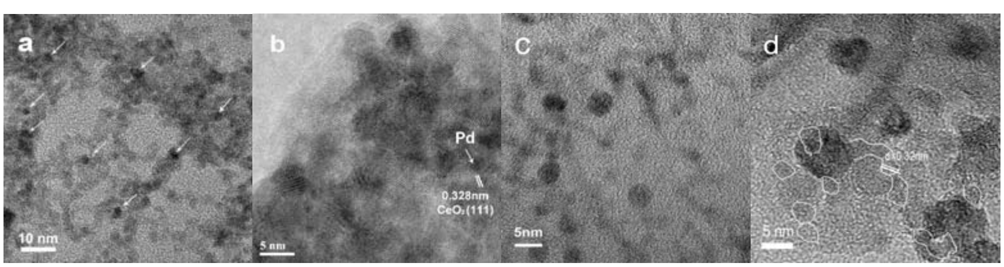
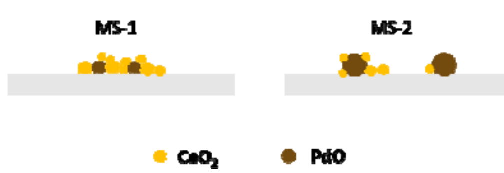


Figure 1. TEM images of as-prepared MS-1(a), MS-2 (c) and the corresponding supported samples: MS-1/Al_2O_3 (b), MS-2/Al_2O_3 (d). The white arrows in (a) indicate the Pd NPs.



Scheme 1. Schematic illustration of MS-1 and MS-2 supported on Al_2O_3 and calcined at 550 °C.

We applied these two model catalysts in CO oxidation reaction and methane combustion reaction. The detailed results are shown in Table 1. The experiment results clearly demonstrated that the specific nano-structure prominently influenced the catalytic performances. For CO oxidation the better activity of MS-1 could be attributed to the more Pd-CeO_2 interface area; which showed lower catalytic methane combustion compared with the PdO species.

Table 1. Catalytic activities of MS-1 and MS-2 in CO oxidation and CH_4 combustion reactions

Samples (Reaction)	E_a (KJ/mol)	TOF ($*10^{-3}$)
MS-1 (CO oxidation)	47.0	5.86 (38 °C)
MS-2 (CO oxidation)	65.1	5.37 (80 °C)
MS-1 (CH_4 combustion)	87.4	2.85 (300 °C)
MS-2 (CH_4 combustion)	99.7	7.38 (300 °C)

3. Conclusions

In this Abstract we have successfully prepared two different types of Pd-CeO_2 nanostructures and supported them on Al_2O_3. Two model reactions, CO oxidation and CH_4 reaction were applied to evaluate the catalytic activities of the nanocatalysts. From the structure and catalytic activity relationship we can observe that the Pd-CeO_2 interface could contribute to the CO oxidation process while PdO with minor ceria decoration would improve the CH_4 combustion reaction.

References

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