Effect of Hydrogen Addition on Catalytic Methane Oxidation at Low Temperature

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Introduction
Catalytic methane oxidation has attracted considerable research interest in energy generation and production of more valuable chemicals, such as methanol and acetic acid [1]. Pd and Pt nanoparticle catalysts have been most often used for their higher activity than other metals in the reaction. However even with Pd and Pt, high reaction temperature is still required to activate methane oxidation. For example, the onset temperature of methane oxidation using Pd and Pt catalyst is 530 and 600 K, respectively [2]. Because a decrease in the reaction temperature has apparent advantages in improving energy utilization efficiency and better controlling the reaction extent, catalytic methane oxidation at low temperature is desired.

Here, we report low temperature catalytic methane oxidation on Pt nanoparticle catalyst by hydrogen addition to the reaction system. The onset temperature of the reaction can be pushed all the way down to room temperature ranges. The effect of hydrogen addition on methane oxidation, as well the behind mechanism, were investigated.

Materials and Methods
Preparation of Pt nanoparticle catalyst: Pt nanoparticles were first synthesized by reducing Pt acetylacetonate in a mixture of 1,2-hexadecanediol, oleylamine, oleic acid and diphenyl ether. The as-synthesized Pt nanoparticles were then put on an alumina support and thermally treated for removing residual organics, with the final product as Pd/Al₂O₃ nanoparticle catalyst.

Characterization: TEM and HRTEM, EDX, and XRD were used to characterize the Pt/Al₂O₃ nanoparticle catalyst.

Catalyst testing: The methane oxidation experiments using the Pt/Al₂O₃ catalyst were carried out in a packed bed reactor (PBR), with an entrance volumetric flow rate of 100 sccm and the feeding gas composition being varied.

Results and Discussion
Figure 1 shows the conversions of O₂ for methane oxidation (X₀₂) at different reaction temperature and with various gas compositions. When 60 vol.% CH₄ and 0.5 vol.% O₂ were used, an onset temperature of 160 °C was observed for the reaction. An 80 °C decrease in the onset temperature was observed when 1.0% H₂ was co-fed, suggesting significant effect of H₂ addition on catalytic methane oxidation. Mechanistic studies were conducted, with possible mechanism being discussed and proposed.

Figure 1. O₂ conversion (X₀₂)-Temperature (T) profiles for catalytic CH₄ oxidation using Pt/Al₂O₃ catalyst and with different gas mixture, which had a volumetric flow rate of 100 sccm and was balanced with Ar.

Significance
In our knowledge, it is the first report that the onset temperature for gas-phase methane oxidation can be pushed to the room temperature range with hydrogen addition. The decrease in the reaction temperature can highly improve the efficiency of energy utilization.

References