The durability of alumina supported Pd catalysts for the combustion of methane in the presence of SO$_2$

In the present work, the 1 wt% Pd/γ-Al$_2$O$_3$, 1 wt% Pd/10 wt% CeO$_2$/γ-Al$_2$O$_3$ and 1 wt% Pd/10 wt% Ce$_{0.8}$Zr$_{0.4}$O$_2$/γ-Al$_2$O$_3$ catalysts were prepared and used for the catalytic combustion of methane. The durability of the catalysts were investigated based on the phenomena of SO$_2$ poisoning in methane catalytic combustion. The presence of SO$_2$ in the reaction gases resulted in a 70-250 °C increase of light-off temperature for methane conversion for all tested catalysts. Pre-treatment of the catalysts with SO$_2$ also led to a decrease in catalytic activity to some extent. The sulphur poisoning and sulphate formation were mainly responsible for the catalyst deactivation. The PdO species contributed to the formation of sulphates by oxidizing SO$_2$ to SO$_3$ species. The sulphates were formed below 600 °C and decomposed above 600-700 °C in the reactions over the catalysts, which led to the inhibition and recovery of the catalytic activity, respectively. The effluent gases (SO$_2$, SO$_3$, and O$_2$) of the SO$_2$-TPD experiments were monitored on-line by a mass spectrometer (Hiden HPR-20 QIC). The results indicated that the introduction of CeO$_2$ or Ce$_{0.8}$Zr$_{0.4}$O$_2$ decreased the decomposition temperature of sulphates by 50-100 °C. Sulphur accumulation on the catalyst surface was investigated by sulphur content analysis and TG measurements after pre-treatment with SO$_2$. The saturated sulphur contents were all about 5 wt% for the three catalysts. In spite of sulphur poisoning, no obvious changes in the particle morphologies or dimensions were observed for the fresh and used catalysts. The introduction of CeO$_2$ or Ce$_{0.8}$Zr$_{0.4}$O$_2$ decreased the decomposition temperature of sulphates by 50-100 °C, indicating that the durability of the catalysts studied in this work could be satisfied for the catalytic combustion of methane that usually takes place above 800 °C.
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