

介孔 Al_2O_3 负载 PdO 催化甲烷燃烧反应性能

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摘要 采用浸渍法制备了介孔 Al_2O_3 (M- Al_2O_3) 负载 PdO 催化剂, 考察了其催化 CH_4 燃烧反应性能。结果表明, 以 M- Al_2O_3 为载体的 PdO 催化剂活性比普通 Al_2O_3 载体高得多, 这很可能与 M- Al_2O_3 的孔道结构对 PdO 物种的限域作用有关。随着 PdO/M- Al_2O_3 催化剂焙烧温度的升高, 甲烷催化燃烧活性先增加后降低, 其中 700 $^{\circ}\text{C}$ 焙烧的催化剂活性最高, 400 $^{\circ}\text{C}$ 反应时 CH_4 转化率为 91%。此时 Pd 物种主要以 PdO 颗粒形式高度分散在载体的介孔孔道内, 而高温焙烧时, Pd 物种主要以 Pd 和 PdO 的混合晶相存在。尽管 900 $^{\circ}\text{C}$ 焙烧制得的催化剂上 CH_4 的转化率降低, 但 TOF 值最大, 这可能与该催化剂中同时存在金属 Pd 和 PdO 有关。

关键词: 介孔氧化铝 钯 甲烷 催化燃烧 活性位

Abstract: Pd catalysts supported on mesoporous alumina (M- Al_2O_3) were prepared by an impregnation method and tested for CH_4 catalytic combustion. PdO/M- Al_2O_3 was more active than the PdO catalyst supported on conventional Al_2O_3 because of a higher dispersion of PdO species induced by the mesoporous structure of M- Al_2O_3 . Moreover, with increasing calcination temperature, the conversion of CH_4 over PdO/M- Al_2O_3 catalysts first increased and then decreased. The best catalytic performance was obtained on the PdO/M- Al_2O_3 catalyst calcined at 700 $^{\circ}\text{C}$, with a CH_4 conversion of 91% at 400 $^{\circ}\text{C}$. Pd species were mainly in mesoporous channels of the support in the form of highly dispersed PdO particles. When calcined at higher temperature, Pd species were in forms of crystalline Pd and PdO. A decline of reactivity and a higher turnover frequency were obtained on the PdO/M- Al_2O_3 catalyst calcined at 900 $^{\circ}\text{C}$, which might be due to the co-existence of metallic Pd and PdO in the catalyst.

Keywords: mesoporous alumina, palladium, methane, catalytic combustion, active site

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