行政院原子能委員會
委託研究計畫研究報告

富氫氣體中一氧化碳抑低方法研究

Removing Carbon Monoxide in Hydrogen Rich Stream

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中文摘要

本研究以懸浮/共沈澱法將氧化鋁引入 $Ce_{1-x}Zr_xO_2$ 固溶液中形成 $Ce_{1-x}Zr_xO_2/Al_2O_3$ 混合氧化物擔體，以臨濕含浸法製備 $CuO/Ce_{1-x}Zr_xO_2/Al_2O_3$ 觸媒，詳細鑑定並應用於富氫進料中之 CO 選擇性氧化反應。引入 Al_2O_3 能增加 $Ce_{1-x}Zr_xO_2$ 的分散性，但不會改變 $Ce_{1-x}Zr_xO_2$ 的主結構也不會減弱 $Ce_{1-x}Zr_xO_2$ 的氧化還原特性，然而引入過量 Al_2O_3 會降低 $CuO/Ce_{1-x}Zr_xO_2/Al_2O_3$ 觸媒中 CuO 與 $Ce_{1-x}Zr_xO_2$ 的緊密接觸界面，降低 CO 的選擇性氧化。適量鋯引入 CeO_2 中能增加晶格氧的移動性，促進 $7\%CuO/Ce_{0.9}Zr_{0.1}O_2/Al_2O_3(20\%)$觸媒之 CO 選擇性氧化反應活性。最佳觸媒 $7\%CuO/Ce_{0.9}Zr_{0.1}O_2/Al_2O_3(20\%)$轉化率達 100%的溫度($T_{100}$)比 $7\%CuO/CeO_2/Al_2O_3$ 觸媒降低約 10℃，且選擇率仍維持近 100%。進料中含 CO_2 及 H_2O 雖會降低 $7\%CuO/Ce_{0.9}Zr_{0.1}O_2/Al_2O_3(20\%)$觸媒活性，但仍能維持高達 90%以上之選擇率及良好的穩定性。$7\%CuO/Ce_{0.9}Zr_{0.1}O_2/Al_2O_3(20\%)$觸媒不僅具有與貴金屬觸媒 $5\%Pt/Al_2O_3$ 相當的良好活性，選擇率更遠高於 $5\%Pt/Al_2O_3$。

Abstract

In this study, alumina was incorporated with the solid solutions of $Ce_{1-x}Zr_xO_2$ to form the supports of $Ce_{1-x}Zr_xO_2/Al_2O_3$ mixed oxides by the suspension/co-precipitation method, and the $CuO/Ce_{1-x}Zr_xO_2/Al_2O_3$ catalysts were prepared by the incipient impregnation. They were characterized and used in the selective oxidation of CO in hydrogen-rich feed. Incorporating Al_2O_3 could increase the dispersion of $Ce_{1-x}Zr_xO_2$, but did not change the main structures of $Ce_{1-x}Zr_xO_2$ and did not weaken their redox properties. Nevertheless, incorporating excess Al_2O_3 would reduce the interfacial intimate contact of CuO clusters with $Ce_{1-x}Zr_xO_2$ for $CuO/Ce_{1-x}Zr_xO_2/Al_2O_3$ catalysts and weakened the selective oxidation of CO. An appropriate amount of zirconium incorporated into CeO_2 increased the mobility of lattice oxygen and enhanced the activity of the $7\%CuO/Ce_{0.9}Zr_{0.1}O_2/Al_2O_3$ catalyst in the selective oxidation of CO. The temperature for complete conversion (T_{100}) of the optimal catalyst $7\%CuO/Ce_{0.9}Zr_{0.1}O_2/Al_2O_3(20\%)$ was about ten degrees lower than that of $7\%CuO/CeO_2/Al_2O_3(20\%)$, and the selectivity achieved was nearly 100%. The activity of the $7\%CuO/Ce_{0.9}Zr_{0.1}O_2/Al_2O_3(20\%)$ catalyst was weakened in the feed in the presence of CO_2 and H_2O, but a selectivity above 90% and a good stability of the catalyst were still maintained. The activity of $7\%CuO/Ce_{0.9}Zr_{0.1}O_2$ $Al_2O_3(20\%)$ catalyst was comparable with, and its selectivity was much larger than, those of the noble catalyst $5\%Pt/Al_2O_3$.