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铜锆复合氧化物的结构对其催化选 择还原－氧化氮性 能的影响

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摘要 用浸渍法和共沉淀法分别制得CuO---ZrO~2复合氧化具物有不同的选择还原NO~x的催化性能，采用XRD，BET，EXAFS和H~2---TPR等手段对样品进行了表征，发现浸渍法制备的样品具有的比表面较大，氧化锆被稳定在四方相。EXAFS实验表明，
浸渍法制得样品的铜离子填入氧化锆表面空穴中，并以Cu^2+形式存在；500℃焙条件下用共沉淀法引入的铜离子可部分取代锆离子，在氧化锆体相高度分散形成均匀的无定形固溶体，
铜离子在氧化锆体相的高度分散是形成表面弧立铜物种的关键。溶入氧化锆体相的铜离子在取代部位由于局部负电荷而使氧化性降低，是共沉淀法制备样品具有较高催化活性的主要原因。

关键词 氧化铜 氧化锆 一氧化氮 催化性能 X射线衍射分析 程序升温还原

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The structure of CuO---ZrO~2mixed oxides and its infiuence on the catalytic selective reduction of NO by propene

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Abstract Different catalytic performances of CuO---ZrO~2 mixed oxides prepared by impregnation or co-precipitiation method for selective reduction of NO(in the presence of O~2) were invstigated. The sturctures and properties of the catalysts were characterized by XRD, BET surface area measurement, EXAFS and H~2 ---TPR measurements. The CuO---ZrO~2 mixed oxides prepared by impregnation method had larger surface area than those prepared by c0- precipitation methos. The zirconium oxide was stabilized at the tetragonal phase. EXAFS measurement shows that in the CuO---ZrO~2 mixed oxides prepared by impregnation method the copper ions first fill in the syrface hole of zirconium oxide and stay there as cu^+. Under the calcination at 500℃ , the Cu^2+ ions that brought into ZeO~2 by co-precipitation method can replace Zr^4+ and form a uniform amorphous solid solution. The high -dispersion of Cu ions in zirconium oxide was the element that led to formation of highly dispersed isolated copper species on the surface. Substitution of Zr^ 4+ by Cu^2+ ions aould decrease the reducibility of copper species and the activity of the complete oxidation of propene. The local structure difference of copper species was the main reason for the high activity and selectivity of the CuO---ZrO~2 mixed oxides prepared by c0-precipitation method.

Key words COPPER OXIDE ZIRCONIUM OXIDE NITROGEN MONOXIDE CATALYTIC BEHAVIOUR X-RAY DIFFRACTION ANALYSIS TEMPERATURE PROGRAMMED REDUCTION

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