

改性钛锆复合氧化物催化环己酮肟气相Beckmann重排反应

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摘要 制备了经 SO_4^{2-} 、 PO_4^{3-} 、 MoO_3 、 WO_3 和 B_2O_3 改性的钛锆复合氧化物(TiO_2 - ZrO_2)催化剂,并用常压连续流动固定床微型反应器考察了它们对环己酮肟气相Beckmann重排反应制己内酰胺的催化性能。结果表明,在反应温度 300°C 时, $\text{B}_2\text{O}_3/\text{TiO}_2$ - ZrO_2 具有较高的己内酰胺选择性(97.0%)和收率(96.7%),且明显高于 $\text{B}_2\text{O}_3/\text{TiO}_2$ 和 $\text{B}_2\text{O}_3/\text{ZrO}_2$ 催化剂,用 N_2 吸附和 NH_3 -TPD, CO_2 -TPD等方法对催化剂的比表面积、孔容、孔分布和表面酸、碱性分别进行了测定,讨论了这些物化性能对催化剂活性、选择性和稳定性的影响。

关键词 [钛锆复合氧化物](#) [催化活性](#) [环己酮肟](#) [气相反应](#) [钛化合物](#) [锆化合物](#) [氧化硼](#) [己内酰胺](#) [贝克曼重排](#)

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Vapor-phase beckmann rearrangement of cyclohexanone oxime over modified titania-zirconia catalyst

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Abstract The vapor-phase Beckmann rearrangement of cyclohexanone oxime to ϵ -caprolactam was examined over a variety of TiO_2 - ZrO_2 mixed oxides modified by addition of sulfate, phosphate, and oxides of molybdenum, tungsten as well as boron. The catalytic reactions were performed in a continuous flow fixed-bed micro-reactor under atmospheric pressure. Boron oxide (B_2O_3) modified TiO_2 - ZrO_2 ($\text{B}_2\text{O}_3/\text{TiO}_2$ - ZrO_2) showed the highest selectivity (97.0%) and yield (96.7%) of ϵ -caprolactam among the catalysts examined. These catalysts were characterized by nitrogen adsorption and temperature-programmed desorption of ammonia and carbon dioxide techniques. The effect of the physico-chemical properties of these catalysts on their catalytic performance was examined. The catalytic properties of $\text{B}_2\text{O}_3/\text{TiO}_2$ - ZrO_2 were better than those of $\text{B}_2\text{O}_3/\text{TiO}_2$ and $\text{B}_2\text{O}_3/\text{ZrO}_2$.

Key words [CATALYTIC ACTIVITY](#) [CYCLOHEXANONE-OXIME](#) [GAS PHASE REACTION](#) [TITANIUM COMPOUNDS](#) [ZIRCONIUM COMPOUNDS](#) [BORON OXIDE](#) [CAPROLACTAM](#) [BECKMANN REARRANGEMENT](#)

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