

K改性Ni-Co-Al三元复合氧化物催化分解N₂O

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Catalytic decomposition of N₂O over potassium promoted Ni-Co-Al ternary mixed oxides

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摘要 制备了(Ni+Co)/Al=3、Ni/Co=0.2(原子比)的NiCoAl三元类水滑石样品,焙烧获得NiCoAl复合氧化物,表面浸渍K₂CO₃溶液制备了K改性催化剂,用于N₂O分解反应,考察了K负载量、焙烧温度等制备参数和O₂、H₂O等反应气氛对催化剂活性的影响。用BET、XRD、H₂-TPR、XPS等技术表征了催化剂的组成结构。结果表明,K的表面改性提高了催化剂对N₂O分解反应的催化活性,其中,400℃预焙烧NiCoAl类水滑石制得复合氧化物,初湿浸渍K₂CO₃溶液,K的负载量为K/(Ni+Co)=0.05,400℃再焙烧制备的催化剂活性较高,有氧有水条件下500℃反应时N₂O可完全分解;在NiCoAl复合氧化物表面负载K₂CO₃组分,降低了催化剂表面Co、Ni元素的电子结合能,弱化了表面Co-O、Ni-O化学键,从而提高了催化剂活性和抗水性。

关键词: N₂O催化分解 K改性NiCoAl复合氧化物 催化剂制备参数 反应气氛 催化活性

Abstract: NiCoAl ternary mixed oxides derived from hydrotalcite-like compounds (HLC) with (Ni+Co)/Al atomic ratio of 3 and Ni/Co of 0.2 were incipiently impregnated by potassium carbonate solution to prepare K-promoted NiCoAl mixed oxides for catalytic decomposition of N₂O. The effects of catalyst preparation parameters such as K loadings, calcination temperatures, and reaction atmospheres on catalytic activity were investigated. The catalysts were characterized by BET, XRD, H₂-TPR, and XPS techniques. The results show that the catalytic activity of NiCoAl mixed oxides is largely enhanced by the addition of K species in the presence of oxygen and steam, and the optimal catalyst preparation parameters are K/(Ni+Co) atomic ratio of 0.05 and calcination temperature of 400℃. XPS and H₂-TPR data show that the electron binding energy of active Co₃O₄ and NiO species over K-promoted catalysts surface shifted to lower value, indicating the surface Co-O and Ni-O bonds were weakened, thus the reduction peaks of Co³⁺ to Co²⁺ and Ni²⁺ to Ni⁰ moved to lower temperature, and the catalytic activity and resistance towards water is improved.

Key words: N₂O catalytic decomposition K-promoted NiCoAl mixed oxides catalyst preparation parameters reaction atmosphere catalytic activity

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