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研究论文

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Nitrous Oxide Decomposition over Alkali-Promoted Magnesium Cobaltite Catalysts

Bahaa M. ABU-ZIED*

Chemistry Department, Faculty of Science, Assiut University, 71516 Assiut, Egypt

Bahaa M. ABU-ZIED*

Chemistry Department, Faculty of Science, Assiut University, 71516 Assiut, Egypt

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摘要 The direct decomposition of N₂O was investigated over a series of magnesium cobaltite catalysts, Mg_xCo_{1-x}Co₂O₄ (0.0 $\leq x \leq$ 1.0), which were prepared by the thermal decomposition of stoichiometric amounts of magnesium hydroxide and cobalt acetate. The thermal genesis of the different catalysts from their precursors was explored using thermogravimetric analysis, differential thermal analysis, and X-ray diffraction. Texture analysis was carried out using N₂ adsorption at -196° C. We found that all the catalysts that were calcined at 500 $^{\circ}$ C have a spinel structure. N₂O decomposition activity was found to increase with an increase in the spinel structure' s magnesium content. The influence of alkali cation promoters (Li, Na, K, and Cs) on the activity of the most active catalyst in the Mg_xCo_{1-x}Co₂O₄ series, i.e. MgCo₂O₄, was also investigated. The sequence of the promotional effect was found to be: un-promoted < Li < Na < Cs < K-promoted catalyst. The reason for the increase in activity on the K/Co ratio was also determined. The highest activity was obtained for the catalyst with a K/Co ratio of 0.05. A continuous decrease in activity was obtained for higher K/Co ratios. This decrease in activity was attributed to the elimination of mesoporosity in the catalysts with K/Co ratios > 0.05, based on N₂ adsorption and scanning electron microscopy results.

关键词: nitrous oxide cobalt oxide magnesium cobaltite spinel alkali promotion

Abstract: The direct decomposition of N₂O was investigated over a series of magnesium cobaltite catalysts, Mg_xCo_{1-x}Co₂O₄ ($0.0 \le x \le 1.0$), which were prepared by the thermal decomposition of stoichiometric amounts of magnesium hydroxide and cobalt acetate. The thermal genesis of the different catalysts from their precursors was explored using thermogravimetric analysis, differential thermal analysis, and X-ray diffraction. Texture analysis was carried out using N₂ adsorption at -196° C. We found that all the catalysts that were calcined at 500° C have a spinel structure. N₂O decomposition activity was found to increase with an increase in the spinel structure' s magnesium content. The influence of alkali cation promoters (Li, Na, K, and Cs) on the activity of the most active catalyst in the Mg_xCo_{1-x}Co₂O₄ series, i.e. MgCo₂O₄, was also investigated. The sequence of the promotional effect was found to be: un-promoted < Li < Na < Cs < K-promoted catalyst. The reason for the increase in activity for the added alkali cations was electronic in nature. Additionally, the dependence of the activity on the K/Co ratio was also determined. The highest activity was obtained for the catalyst with a K/Co ratio of 0.05. A continuous decrease in activity was obtained for higher K/Co ratios. This decrease in activity was attributed to the elimination of mesoporosity in the catalysts with K/Co ratios > 0.05, based on N₂ adsorption and scanning electron microscopy results.

Keywords: nitrous oxide, cobalt oxide, magnesium cobaltite, spinel, alkali promotion

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