

ZnCr基催化剂煅烧温度对异丁醇合成性能的影响

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Effect of calcination temperature on the performance of ZnCr based catalyst in isobutanol synthesis

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摘要 研究了煅烧温度对ZnCr基催化剂合成异丁醇性能的影响。结果表明,随着煅烧温度的升高,催化剂的活性和产物分布都发生了较大的变化。催化剂在较低的温度下煅烧,液相产物中醇主要是甲醇和异丁醇;在较高的温度下煅烧,液相产物醇的分布符合A-S-F方程。用BET、XRD、H₂-TPR、XPS等技术手段对催化剂结构参数、体相结构、还原性能、表面组成进行表征。结果表明,在300℃煅烧时,催化剂中的ZnO和Cr₂O₃未完全形成非计量尖晶石Zn_xCr_{2/3(1-x)}O;400℃煅烧时,催化剂中形成了最多量非计量尖晶石Zn_xCr_{2/3(1-x)}O;当煅烧温度高于400℃时,随着煅烧温度进一步升高,非计量尖晶石Zn_xCr_{2/3(1-x)}O逐步发生了分解,生成了更多量的ZnO和Cr₂O₃,导致催化剂的活性随之下降。进一步证明了非计量尖晶石Zn_xCr_{2/3(1-x)}O是该催化反应活性相。

关键词: ZnCr基催化剂 CO加氢 异丁醇 合成气 非计量尖晶石Zn_xCr_{2/3(1-x)}O

Abstract: The effect of calcination temperature on the performance of ZnCr based catalysts in isobutanol synthesis was investigated; the texture properties, bulk structure, reducibility and surface composition of the catalysts were characterized by BET, XRD, H₂-TPR and XPS. The results indicate that both the activity and product selectivity of the ZnCr catalyst are greatly influenced by its calcination temperature. The catalyst calcined at low temperature shows high selectivity to methanol and isobutanol, while the product distribution over the catalyst calcined at high temperature obeys the A-S-F equation. Calcination at 300℃ is insufficient to get a complete formation of non-stoichiometric spinel Zn_xCr_{2/3(1-x)}O, while calcination at 400℃ gives the maximum amount of non-stoichiometric spinel Zn_xCr_{2/3(1-x)}O in the ZnCr based catalyst; however, further increasing the calcination temperature may cause the decomposition of certain non-stoichiometric spinel Zn_xCr_{2/3(1-x)}O to ZnO and Cr₂O₃, which will reduce its catalytic activity in isobutanol synthesis. Such results suggest that non-stoichiometric spinel Zn_xCr_{2/3(1-x)}O is possibly the active phase of the ZnCr based catalyst in isobutanol synthesis.

Key words: ZnCr based catalyst carbon monoxide hydrogenation isobutanol synthesis syngas non-stoichiometric spinel Zn_xCr_{2/3(1-x)}O

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