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制备方法对 Ni/ZnO 催化丙三醇重整-氢解性能的影响

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摘要 采用浸渍法、共沉淀法、水热法和碳微球硬模板法制备了 Ni/ZnO 催化剂, 运用 X 射线衍射、程序升温还原、透射电子显微镜和氢滴定等手段对其进行了表征, 并用于连续固定床反应器中无外加氢气条件下的丙三醇重整-氢解反应. 结果表明, 在较低空速下, 生成的 1,2-丙二醇 (1,2-PDO) 易在 Ni 分散度较高的催化剂上进一步裂解为乙醇和气相产物; 而在较高空速下, 其选择性受制于中间产物丙酮醇的加氢. 在优化的空速下, Ni 分散度越高越有利于 1,2-PDO 的生成. 在 Ni 分散度最高的 Ni/ZnO 催化剂上, 当丙三醇质量空速为 0.84 h－1 时, 1,2-PDO 选择性最高, 为 54.9%, 丙三醇转化率为 85.4%.

关键词: 镍 氧化锌 丙三醇 重整 氢解 1,2-丙二醇 分散度

Abstract: The catalytic conversion of glycerol to 1,2-propanediol (1,2-PDO) is generally conducted batch-wise in an autoclave in the presence of high pressure H_2. The reforming and hydrogenolysis of glycerol to 1,2-PDO over Ni/ZnO catalysts in a continuous flow fixed-bed reactor without added H_2 was reported. The Ni/ZnO catalysts were prepared by wetness impregnation (WI), co-precipitation (CP), hydrothermal treatment (HT), and carbon microsphere hard-templating (CT) methods. The catalysts were characterized by X-ray diffraction (XRD), temperature-programmed reduction (TPR), transmission electron microscopy (TEM), and H_2 titration. At a low weight hourly space velocity (WHSV) of glycerol, the 1,2-PDO produced got degraded to ethanol and gas phase products over catalysts with high Ni dispersion, while at a high WHSV, the selectivity for 1,2-PDO was limited by the hydrogenation of the acetol intermediate. At the optimized WHSV, the catalyst with a higher Ni dispersion was more selective for 1,2-PDO, and over the Ni/ZnO catalyst with the highest Ni dispersion, the highest selectivity of 54.9% for 1,2-PDO was obtained at a glycerol conversion of 85.4% at the WHSV of 0.84 h^{-1}.

Keywords: nickel, zinc oxide, glycerol, reforming, hydrogenolysis, 1,2-propanediol, dispersion degree

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