化学学报 1998 56 (3): 215-222 ISSN: 0567-7351 CN: 31-1320/06

中温镍水蒸汽转化催化剂积碳动力学与机理研究

关毅,秦永宁,张鉴

天津大学化学工程系

收稿日期 修回日期 网络版发布日期 接受日期

摘要 研究了833-948K和常压下, C7H16在镍催化剂上进行水蒸汽转化过程中的积碳反应动力学, 获得了相应的动力学方程。并探讨了相应的反应机理, 提出在镍表面同时进行的水蒸汽转化和积碳反应是两个相对独立的反应。指出在较高温度下, 水蒸汽会从简单非解离吸附转变为解离吸附, 这一变化不仅改变了反应级数, 更重要的是它提高了水蒸汽参与表面反应的能力, 特别是加强了氢对积碳反应的作用; 同时庚烷的吸附热也随之产生相当大变化。分析了出现在本温度区内积碳反应负表观活化能的来源。

关键词 反应机理 催化剂 镍 反应动力学 甲苯 活化能 结碳

分类号 0643

Study on the coking kinetics and mechanism on steam reforming Ni-catalyst

GUAN YI,QIN YONGNING,ZHANG LIU

Abstract The kinetics of carbon deposition has been investigated on a supported nickel catalyst for steam reforming of naphtha and the mechanism has been described. The following experimental conditions were applied: 1.01×10^5Pa and 823-948K. Empirical rate equations of carbon deposition with H2O were obtained while empirical rate equations of carbon deposition without H2O were also given only for comparison. In the experimental temperature range, the deep dehydrogenation of adsorbed naphtha is still suggested as the rate limiting step of carbon deposition reaction, but the study revealed that H2O on Ni-surface was dissociatively adsorbed. As a result, the order of steam partial pressure changes from -1 to -0.62 and the effect of hydrogen on carbon deposition increases at the same time. The analysis of mechanism further explains why H2 can speed the carbon deposition. So far as the reason for the formation of apparent negative activation energy is concerned, it has been argued about for a long time, but no agreement has been made yet. The observations are shown to be best explained with the cooperative effects including pyrolysis reaction, adsorption and steam reforming reaction.

Key words REACTION MECHANISM CATALYST NICKEL REACTION KINETICS METHYLBENZENE ACTIVATION ENERGY

DOI:

通讯作者