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硅烷处理N_2O氧化苯生产苯酚催化剂抑制结碳失活研究

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摘要 The main cause to the deactivation of ZSM-5 catalyst, used for oxidation of benzene to phenol (BTOP) by nitrous oxide, is that the carbon deposition on the catalyst surface blocks the mouth of pores of the catalyst. In the experiments, ZSM-5 catalyst was modified by chemical surface deposition of silicon, and then the effect of modification condition on the catalyst activation was studied. The catalyst samples were characterized by XRF, EPS, XRD, TEM, N2 adsorption at low temperature, pyridine adsorption-infrared technique and etc. All the above results show that the uniform SiO2 membrane can be formed on ZSM-5 crystal surface. The SiO2 membrane covers the acid centers on ZSM-5 surface to inhibit surface coking, to avoid or decrease the possibility of ZSM-5 pore blockage so that the catalyst activity and stability can be improved efficiently. The optimum siliconiting conditions determined by the experiments are as follows: 4% load of silanizing agent, volume (ml)/mass (g) ratio of hexane/ZSM-5=15/1, and 16 h of modification time. Compared with the samples without siliconiting treatment, the samples treated under the above optimum condition can increase the productivity of phenol by 14% for 3h reaction time and by 41% for 6 h reaction time respectively.

关键词 石碳酸 一氧化二氮 硅烷化反应 改性技术 催化剂 钝化作用

分类号

Coking and Deactivation of Catalyst Inhibited by Silanization Modification in Oxidation of Benzene to Phenol with Nitrous Oxide

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Abstract

The main cause to the deactivation of ZSM-5 catalyst, used for oxidation of benzene to phenol (BTOP) by nitrous oxide, is that the carbon deposition on the catalyst surface blocks the mouth of pores of the catalyst. In the experiments, ZSM-5 catalyst was modified by chemical surface deposition of silicon, and then the effect of modification condition on the catalyst activation was studied. The catalyst samples were characterized by XRF, EPS, XRD, TEM, N2 adsorption at low temperature, pyridine adsorption-infrared technique and etc. All the above results show that the uniform SiO2 membrane can be formed on ZSM-5 crystal surface. The SiO2 membrane covers the acid centers on ZSM-5 surface to inhibit surface coking, to avoid or decrease the possibility of ZSM-5 pore blockage so that the catalyst activity and stability can be improved efficiently. The optimum siliconiting conditions determined by the experiments are as follows: 4% load of silanizing agent, volume (ml)/mass (g) ratio of hexane/ZSM-5=15/1, and 16 h of modification time. Compared with the samples without siliconiting treatment, the samples treated under the above optimum condition can increase the productivity of phenol by 14% for 3h reaction time and by 41% for 6 h reaction time respectively.

Key words nitrous oxide phenol oxidation of benzene to phenol (BTOP) reaction coking deactivation silanization SiO2 membrane

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