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论文

载体酸性对铜基交联黏土催化剂上C_3H_6选择性催化还原NO反应的影响

林绮纯[1], 林维明[2], 郝吉明[1], 李俊华[1], 傅立新[1]

1. 清华大学环境科学与工程系, 北京100084; 2. 广州大学化工系, 广州 510405

摘要:

采用聚合羟基复合阳离子合成交联蒙脱土Al-Ce-PILC, 并分别用NH_4NO_3和$(NH_4)_2SO_4$处理后, 将其作为载体, 采用浸渍法制备了应用于C3H6选择性催化还原NO反应的铜基交联黏土催化剂Cu/Al-Ce-PILC. 用Py-IR, IR和DSC等表征技术研究了不同处理方式对Al-Ce-PILC的结构、 酸性和催化剂活性的影响. 结果表明, 未经处理的Al-Ce-PILC中同时存在L酸和B酸, 以L酸为主, Cu/Al-Ce-PILC上NO的最大转化率仅为18。5%; 用NH_4NO_3处理提高了L酸量, NO转化率降低; 而用$(NH_4)_2SO_4$处理改变了Al-Ce-PILC的酸性结构, 增大了B酸量, 并形成了超强酸中心, 催化剂上NO转化率显著提高, 在350 ℃时达最大值50.2%. B酸对于Cu/Al-Ce-PILC上NO的还原是必要的, 它有利于C_3H_6吸附并适度氧化为活性中间物种, 其酸量和酸强度的增加是催化剂活性改善的主要原因.

关键词: Cu基交联黏土催化剂; 酸性; $(NH_4)_2SO_4$处理; 氮氧化物; 选择性催化还原(SCR)

Effect of Support Acidity on Selective Catalytic Reduction of NO by C_3H_6 over Cu-based Pillared Clay Catalyst

LIN Qi-Chun[1], LIN Wei-Ming[2], HAO Ji-Ming[1], LI Jun-Hua[1], FU Li-Xin[1]

1. Department of Environmental Science and Engineering, Tsinghua University, Beijing 100084, China; 2. Department of Chemical Engineering, Guangzhou University, Guangzhou 510405, China

Abstract:

New pillared clay catalysts were studied for NOx removal by hydrocarbon in the presence of oxygen. The purpose of this work is to study the role of support acidity in the selective catalytic reduction(SCR) of NO by propylene. In this paper, montmorillonite was pillared by multi-oligomeric hydroxyl cation to synthesize Al--Ce-PILC and then treated by NH4NO3 and(NH4)2SO4 respectively. Cu-based pillared clay catalysts(Cu/Al-Ce-PILC) were prepared by impregnation and applied to C-H_6 SCR of NO. The influence of treatment methods on the structure and acidity of Al-Ce-PILC and the performance of Cu/Al-Ce-PILC were characterized by Py-IR,IR and DSC. The results show that, both Lewis and Br-nsted acid sites exist on Al-Ce-PILC, with Lewis acid sites as the main ones, and the maximum NO conversion of Cu/Al-Ce-PILC reaches only 18.5%. As a result of the NH_4NO_3 treatment, the Lewis acid sites increase, while NO conversion decreases. As for Al-Ce-PILC treated by$(NH_4)_2SO_4$, more Br-nsted acid sites are identified and a superacid structure is formed, and the NO conversion over Cu/Al-Ce-PILC catalyst increases remarkably to 50.2% at 350 ℃, which is associated with the change of the support acidity. It is concluded that Br-nsted acid sites are necessary for the SCR reaction of NO because they promote the absorption of C_3H_6 to form proper intermediates that are active for the reduction of NO to N_2. The increase of Br-nsted acid sites plays a vital role in the improvement of catalytic performance.

Keywords: Cu-based pillared clay catalyst; Acidity; $(NH_4)_2SO_4$ treatment; Nitric oxides; Selective catalytic reduction(SCR)

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通讯作者: 林绮纯(1972年出生), 女, 博士, 讲师, 主要从事环境催化的研究. E-mail: lqc@mail.tsinghua.edu.cn

作者简介:

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