燃料化学学报
JOURNAL OF FUEL CHEMISTRY AND TECHNOLOGY
ISSN 0253-2409
CN 14-1140/TQ

燃料化学学报 » 2013, Vol. 41 » Issue (04): 506-512 DOI:

铁铈复合氧化物催化剂SCR脱硝反应动力学研究

熊志波[1,2], 郭东旭[1], 路春美[1,2], 张信莉[1]

1. 山东大学 能源与动力工程学院, 山东 济南 250061;
2. 山东大学 燃煤污染物减排国家工程实验室, 山东 济南 250061

Kinetic study on the selective catalytic reduction of NO with NH_3 over iron-cerium mixed oxide

XIONG Zhi-bo[1,2], GUO Dong-xu[1], LU Chun-mei[1,2], ZHANG Xin-li[1]

1. School of Energy and Power Engineering, Shandong University, Jinan 250061, China;
2. National Engineering Laboratory for Coal-Burning Pollutants Emission Reduction, Shandong University, Jinan 250061, China

全文: PDF (657 KB) HTML (1 KB) 输出: BibTeX | EndNote (RIS) 背景资料

摘要 利用共沉淀法制备了铁铈复合氧化物催化剂,在积分实验系统上考察了NO初始浓度、NH_3/NO比及O_2浓度对其SCR脱硝活性的影响;并借助微分系统探讨了其SCR脱硝的催化反应动力学,构建了铁铈复合氧化物催化剂的催化脱硝反应动力学模型.实验结果表明,NO初始浓度越高,每克催化剂的NO转化率越高;随着NH_3/NO比的增加,NO转化率先迅速增加后趋势减缓,最终趋于稳定;O_2在NH_3-SCR反应中起着重要的作用;在175~225 ℃下,$Fe_{0.95}Ce_{0.05}O_z$催化剂的NO和NH_3反应级数分别为1级和0级,O_2的反应级数接近0.5级,该反应的表观活化能为42.6 kJ/mol.

关键词: 铁铈复合氧化物催化剂 选择性催化还原脱硝 NO 反应动力学

Abstract: Iron-cerium mixed metal oxide ($Fe_{0.95}Ce_{0.05}O_z$) was prepared through co-precipitation method and used as the catalyst in the selective catalytic reduction of NO with NH_3 (NH_3-SCR); the effects of initial concentration of NO, molar ratio of NH_3/NO and O_2 concentration on its NH_3-SCR activity were investigated in an integral reactor at steady-state and the kinetics were considered in a differential reactor. The results indicated that the amount of NO converted over per unit catalyst increases with the initial concentration of NO; with the increase of NH_3/NO mol ratio, NO conversion is firstly increased sharply and then gradually reaches a stable value; O_2 in the flue gas also plays an important role in the NH_3-SCR reaction. The NH_3-SCR reaction over $Fe_{0.95}Ce_{0.05}O_z$ exhibits first-order with respect to NO, zero-order to NH_3, and nearly 0.5-order to O_2 at 175~275 ℃; the apparent activation energy is 42.6 kJ/mol.

Key words: iron-cerium mixed oxide catalyst selective catalytic reduction of NO (SCR) NO kinetics

收稿日期: 2012-11-08;

基金资助:

国家自然科学基金面上项目(51276101); 教育部博士点基金(20120131110022); 山东省自然科学基金面上项目(ZR2012EEM013).

通讯作者: 路春美(1958-), 女, 山东淄博人, 博士, 教授, 研究方向为热能工程和环境工程.E-mail: cml@sdu.edu.cn. E-mail: cml@sdu.edu.cn

服务
把本文推荐给朋友
加入我的书架
加入引用管理器
E-mail Alert
RSS
作者相关文章
熊志波
郭东旭
路春美
张信莉

引用本文:

熊志波,郭东旭,路春美等. 铁铈复合氧化物催化剂SCR脱硝反应动力学研究[J]. 燃料化学学报, 2013, 41(04): 506-512.

XIONG Zhi-bo,GUO Dong-xu,LU Chun-mei et al. Kinetic study on the selective catalytic reduction of NO with NH_3 over iron-cerium mixed oxide[J]. J Fuel Chem Technol, 2013, 41(04): 506-512.

链接本文:

http://rlhxxb.sxicc.ac.cn/CN/ 或 http://rlhxxb.sxicc.ac.cn/CN/Y2013/V41/I04/506

[1] 沈伯雄, 熊丽仙, 刘亭, 王静, 田晓娟. 纳米负载型V_2O_5-WO_3/TiO_2催化剂碱中毒及再生研究[J]. 燃料化学学报, 2010, 38(1): 85-90. (SHEN Bo-xiong, XIONG Li-xian, LIU Ting, WANG Jing, TIAN Xiao-juan. Alkali deactivation and regeneration of nano V_2O_5-WO_3/TiO_2 catalysts[J]. Journal

of Fuel Chemistry and Technology, 2010, 38(1): 85-90.)

[2] 沈伯雄,马娟.SiO_2改性的V_2O_5-WO_3/TiO_2催化剂抗碱中毒性能研究[J].燃料化学学报, 2012, 40(2): 247-251. (SHEN Bo-xiong, MA Juan.Alkali-resistant performance of V_2O_5-WO_3/TiO_2 catalyst modified by SiO_2[J]. Journal of Fuel Chemistry and Technology, 2012, 40(2): 247-251.)

[3] LONG R Q,YANG R T. The promoting role of rare earth oxides on Fe-exchanged TiO_2-pillared clay for selective catalytic reduction of nitric oxide by ammonia[J]. Appl Catal B, 2000, 27(2): 87-95.

[4] LIU F D, HE H, ZHANG C B, SHAN W P, SHI X Y. Mechanism of the selective catalytic reduction of NO_x with NH_3 over environmental-friendly iron titanate catalyst[J]. Catal Today, 2011, 175(1): 18-25.

[5] LIU F D, HE H, ZHANG C B, FENG Z C, ZHENG L R, XIE Y N, HU T D. Selective catalytic reduction of NO with NH_3 over iron titanate catalyst: Catalytic performance and characterization[J]. Appl Catal B, 2010, 96(3-4): 408-420.

[6] PRANIT S M, NELSON S, RACHEL M, VEMURRI B, MICHAEL P H. Selective catalytic reduction of NO with NH_3 on iron zeolite monolithic catalysts:Steady-state and transient kinetics[J]. Appl Catal B: Environ, 2011, 104(1-2): 110-126.

[7] 范红梅, 仲兆平, 金保升, 李峰, 刘涛, 翟俊霞. V_2O_5-WO_3/TiO_2催化剂氨法SCR脱硝反应动力学研究[J].燃料化学学报, 2006, 34(3): 377-380. (FAN Hong-mei, ZHONG Zhao-Ping, JIN Bao-sheng, LI Feng, LIU Tao, ZHAI Jun-xia. Kinetics study of the selective catalytic reduction of NO with NH_3 over V_2O_5-WO_3/TiO_2 catalyst[J]. Journal of Fuel Chemistry and Technology, 2006, 34(3): 377-380.)

[8] HUANG H Y, LONG R Q, YANG R T. Kinetics of selective catalytic reduction of NO with NH_3 on Fe-ZSM-5 catalyst[J]. Appl Catal A, 2002, 235(1-2): 241-251.

[9] LI Z, SHEN L T, HUANG W, XIE K C. Kinetics of selective catalytic reduction of NO by NH_3 on Fe-Mo/ZSM-5 catalyst[J]. J Environ Sci, 2007, 19(12): 1516-1519.

[10] XIONG Z B, LU C M, GUO D X, ZHANG X L, HAN K H. Selective catalytic reduction of NO_x with NH_3 over iron-cerium mixed oxide catalyst: catalytic performance and characterization[J]. J Chem Technol Biot, DOI 10.1002/jctb. 3966.

[11] 熊志波, 路春美. 铁铈复合氧化物催化剂SCR脱硝的改性研究[J]. 燃料化学学报, 2013, 41(3): 361-367. (XIONG Zhi-bo, LU Chun-mei. Study on the modfication of iron-cerium mixed oxide catalyst for selective catadlytic reduction of NO[J]. Journd of Fuel Chemistry and Technology, 2013, 41(3): 361-367..

[12] WU Z B, JIANG B Q, LIU Y, ZHAO W R, GUAN B. H. Experimental study on a low-temperature SCR catalyst based on MnO_x/TiO_2 prepared by sol-gel method[J]. J Hazard Mater, 2007, 145(3): 488-494.

[13] GAO X. JIANG Y, ZHONG Y, LUO Z Y, CEN K. F. The activity and characterization of CeO_2-TiO_2 catalysts prepared by the sol-gel method for selective catalytic reduction of NO with NH_3[J]. J Hazard Mater, 2010, 174(1-3): 734-739.

[14] BALLE P, GEIGER B, KLUKOWSKI D, PIGNATELLI M, WOHNRAU S, MENZEL M, ZIRKWA I, BRUNKLAUS G, KURETI S. Study of the selective catalytic reduction of NO_x on an efficient Fe/HBEA zeolite catalyst for heavy duty diesel engines[J]. Appl Catal B, 2009, 91(3-4): 587-595.

[15] BOSCH H, JANSSEN F J J G, VAN DEN KERKHOF F M G, OLDENZIEL J, VAN OMMEN J G, ROSS J R H. The activity of supported vanadium oxide catalysts for the selective reduction of NO with ammonia[J]. Appl Catal, 1986, 25(1-2): 239-248.

[16] KOEBEL M, ELSENER M, MADIA G. Reaction pathways in the selective catalytic reduction process with NO and NO_2 at low temperature [J]. Ind Eng Chem Res, 2001, 40(1): 52-59.

[1] 王芳, 曾玺, 韩江则, 张聚伟, 刘云义, 汪印, 李寡明, 余剑, 许光文. 微型流化床与热重测定煤焦-CO_2气化反应动力学的对比研究[J]. 燃料化学学报, 2013, 41(04): 407-413.

[2] 熊志波, 路春美. 铁铈复合氧化物催化剂SCR脱硝的改性研究[J]. 燃料化学学报, 2013, 41(03): 361-367.

[3] 刘洪涛, 韩奎华, 路春美, 李辉. O_2/CO_2气氛下木醋调质石灰石再燃/先进再燃脱硝性能研究[J]. 燃料化学学报, 2013, 41(02): 228-234.

[4] 张晓鹏, 沈伯雄. Mn/Ce-ZrO_2催化剂低温NH_3-SCR脱硝性能研究[J]. 燃料化学学报, 2013, 41(01): 123-128.

[5] Mohammad Karimi, Forouzan Aboufazeli, Hamid Reza Lotfi Zadeh Zhad, Omid Sadeghi, Ezzatollah Najafi. Electrocatalytic performance of Pt/Ru/Sn/W fullerene electrode for methanol oxidation in direct methanol fuel cell[J]. 燃料化学学报, 2013, 41(01): 91-95.

[6] 王力, 陈继涛, 张华伟, 李敏, 张林林, 刘珊珊. $KMnO_4$溶液改性半焦对烟气中气态Hg^0的吸附性能研究[J]. 燃料化学学报, 2012, 40(12): 1498-1504.

[7] 彭莉莉, 黄妍, 李建光, 张俊丰. CoO_x-CeO_x/ZrO_2催化氧化NO性能及抗SO_2毒化研究[J]. 燃料化学学报, 2012, 40(11): 1377-1383.

[8] 谢建军, 杨学民, 陈安合, 丁同利, 宋文立, 林伟刚. 煤炭解耦燃烧过程N迁移与转化Ⅲ:多组分气相化学反应实验[J]. 燃料化学学报, 2012, 40(10): 1172-1178.

[9] 李娟, 海航, 闫常峰, 胡蓉蓉, 么志伟, 罗伟民, 郭常青, 李文博. 焙烧温度对二甲醚水蒸气重整制氢 Cu/ZnO/Al_2O_3/Cr_2O_3+H-ZSM-5双功能催化剂性能的影响[J]. 燃料化学学报, 2012, 40(10): 1240-1245.

[10] 任倩茹, 张泽凯, 徐彪, 陈银飞. 负载型钙钛矿$La_{0.8}Sr_{0.2}MnO_3$/SBA-15催化燃烧甲苯的研究[J]. 燃料化学学报, 2012, 40(09): 1142-1146.

[11] 郭锡坤, 李式伟, 郭伟斌. Cu/Dy/$S_2O_8^{2-}$/Al-PILR催化剂制备及其选择催化还原 NO[J]. 燃料化学学报, 2012, 40(09): 1121-1127.

[12] 谢建军, 杨学民, 陈安合, 丁同利, 宋文立, 林伟刚. 煤炭解耦燃烧过程N迁移与转化Ⅱ:单组分气相化学反应实验[J]. 燃料化学学报, 2012, 40(09): 1051-1059.

[13] 杜梅芳, 张影, 樊俊杰, 吴高贺, 侯宁普, 鲁贵林, 张忠孝. 气体再燃低NO_x燃烧中NO与NH_i的反应机理研究[J]. 燃料化学学报, 2012, (08): 935-942.

[14] 杨辉, 刘豪, 周康, 闫志强, 赵然, 刘子红, 柳朝晖, 邱建荣. 活性炭纤维吸附脱除NO过程中NO氧化路径分析[J]. 燃料化学学报, 2012, (08): 1002-1008.

Wan Azelee Wan Abu Bakar, Rusmidah Ali, Abdul Aziz Abdul Kadir, Salmiah Jamal Mat Rosid, Nurul Shafeeqa Mohammad. Catalytic

[15] methanation reaction over alumina supported cobalt oxide doped noble metal oxides for the purification of simulated natural gas[J]. 燃料化学学报, 2012, 40(07): 822-830.