

甲烷在金属铁及氧化铁表面还原NO的研究

苏亚欣, 任立铭, 苏阿龙, 邓文义

东华大学 环境科学与工程学院, 上海 201620

NO reduction by methane on the surface of iron and iron oxides

SU Ya-xin, REN Li-ming, SU A-long, DENG Wen-yi

School of Environmental Science and Engineering, Donghua University, Shanghai 201620, China

- 摘要
- 参考文献
- 相关文章
- 点击分布统计
- 下载分布统计

全文: [PDF](#) (716 KB) [HTML](#) (1 KB) 输出: [BibTeX](#) | [EndNote](#) (RIS) [背景资料](#)

摘要 在程序控温电加热水平陶瓷管反应器、 N_2 气氛和模拟烟气气氛及 $300\sim 1100^\circ C$ 时,对甲烷在金属铁及其氧化铁表面还原NO的特性进行了实验研究。为使甲烷在脱硝反应后完全燃尽以及脱硝反应过程生成的CO等中间产物完全燃尽,在第一段加热炉后串联了第二段加热炉,补充氧气,实现燃尽。结果表明,甲烷在金属铁及氧化铁表面能够高效地还原NO。在 N_2 气氛中,在 $900^\circ C$ 以上温度范围内甲烷在金属铁表面的脱硝效率超过95%,与甲烷在氧化铁表面的脱硝效率差别很小。在模拟烟气条件下,当过量空气系数小于1.0时,在 $900^\circ C$ 以上时,甲烷在金属铁和氧化铁表面的脱硝效率都能超过90%,且未燃尽和燃尽两种条件下NO的还原率相差不大。NO同时通过金属铁的直接还原和甲烷的再燃还原两种反应机理脱除。而甲烷则通过还原氧化铁为金属铁,从而使金属铁直接还原NO可持续进行。同时,甲烷再燃反应的中间产物HCN/ NH_3 等被氧化铁还原,从而使燃尽后的脱硝效率不下降。研究结果表明,甲烷和金属铁或氧化铁在富燃料条件下可有效地还原NO。

关键词: NO还原 甲烷 铁 氧化铁

Abstract: NO reduction by methane on the surface of iron and iron oxides was experimentally investigated in a one-dimensional temperature-programmed ceramic tubular reactor at $300\sim 1100^\circ C$ in both nitrogen and simulated flue gas atmospheres. To ensure that the residual methane after NO reduction and the intermediates (e.g. CO) formed during the NO reduction were completely burned out, a second furnace with a supply of O_2 was connected in series after the first furnace. The results indicated that methane can effectively reduce NO to N_2 over the surface of metallic iron and iron oxides. In N_2 atmosphere, more than 95% of NO is reduced by methane over metallic iron at a temperature above $900^\circ C$, which is very close to that for NO reduction over iron oxides. In the simulated flue gas atmosphere with an excessive air ratio being lower than 1.0, more than 90% of NO is reduced by methane over both metallic iron and iron oxides at a temperature above $900^\circ C$; there is little difference in NO reduction under both burnout or non-burnout conditions. NO is reduced simultaneously via two routes, i.e. the direct reduction by metallic iron and the reduction by reburning of methane. Iron oxides are reduced to metallic iron by methane through partial oxidation over iron oxides to maintain the sustainable reduction of NO by metallic iron. At the same time, the intermediate products during NO reduction by methane such as HCN/ NH_3 are converted by iron oxides, which prevent the NO reduction efficiency from dropping after burnout. The present results then prove that methane can effectively reduce NO over iron or iron oxides under fuel rich condition.

Key words: NO reduction methane iron iron oxides

收稿日期: 2013-03-25;

基金资助:

国家自然科学基金(51278095);上海市自然科学基金(11ZR1401000)。

通讯作者: 苏亚欣(1972-),男,博士,副教授,E-mail:suyx@dhu.edu.cn,Tel:021-67792552。 E-mail:

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

- ▶ 苏亚欣
- ▶ 任立铭
- ▶ 苏阿龙
- ▶ 邓文义











引用本文:

苏亚欣,任立铭,苏阿龙等. 甲烷在金属铁及氧化铁表面还原NO的研究[J]. 燃料化学学报, 2013, 41(11): 1393-1400.

SU Ya-xin, REN Li-ming, SU A-long et al. NO reduction by methane on the surface of iron and iron oxides[J]. J Fuel Chem Technol, 2013, 41(11): 1393-1400.

链接本文:

<http://rlhxxb.sxicc.ac.cn/CN/> 或 <http://rlhxxb.sxicc.ac.cn/CN/Y2013/V41/I11/1393>

- [1] 马凤哪, 程伟琴. 国内火电厂氮氧化物排放现状及控制技术探讨[J]. 广州化工, 2011, 39(15): 57-59. (MA Feng-na, CHENG Wei-qin. The discharge status and controlling measures of nitrogen oxides of thermal power plants in China[J]. Guangzhou Chemical Industry, 2011, 39(15): 57-59.)
- [2] PARVULESCU V I, GRANGE P, DELMON B. Catalytic removal of NO[J]. Catal Today, 1998, 46(4): 233-316. 
- [3] JANSSEN F, MEIJER R. Quality control of DeNO_x catalysts performance testing, surface analysis and characterization of DeNO_x catalysts [J]. Catal Today, 1993, 16(2): 157-185. 
- [4] CENTI G, PERATHONER S. Introduction: State of the art in the development of catalytic processes for the selective catalytic reduction of NO_x into N₂[J]. Stud Surf Sci Catal, 2007, 171: 1-24. 
- [5] BETHKE KA, KUNG M C, YANG B, SHAH M, ALT D, LI C, KUNG H H. Metal oxide catalysts for lean NO_x reduction[J]. Catal Today, 1995, 26(2): 169-183. 
- [6] BETHKE K A, ALT D, KUNG M C. NO reduction by hydrocarbons in an oxidizing atmosphere over transition metal-zirconium mixed oxides [J]. Catal Lett, 1994, 25(1/2): 37-48. 
- [7] ILIOPOULOU E F, EVDUO A P, LEMONIDOU A A, VASALOS I A. Ag/alumina catalysts for the selective catalytic reduction of NO_x using various reductants[J]. Appl Catal A: Gen, 2004, 274(1/2): 179-189. 
- [8] KOTSIFA A, KONDARIDES D I, VERYKIOS X E. A comparative study of the selective catalytic reduction of NO by propylene over supported Pt and Rh catalysts[J]. Appl Catal B: Environ, 2008, 80(3/4): 260-270. 
- [9] LIU Z, WANG K, ZHANG X, WANG J, CAO H, GONG M, CHEN Y. Study on methane selective catalytic reduction of NO on Pt/Ce_{0.67}Zr_{0.33}O₂ and its application[J]. J Nat Gas Chem, 2009, 18(1): 66-70. 
- [10] GRADON B, LASEK J. Investigation of reduction of NO to N₂ by reaction with Fe[J]. Fuel, 2010, 89(11): 3505-3509. 
- [11] 苏亚欣, 苏阿龙, 成豪. 金属铁直接催化还原NO的实验研究[J]. 煤炭学报, 2013, 38(S1): 206-210. (SU Ya-xin, SU A-long, CHENG Hao. Experimental study on direct catalytic reduction of NO by metallic iron[J]. Journal of China Coal Society, 2013, 38(s1): 206-210.)
- [12] 李然家, 沈师孔. 晶格氧用于甲烷氧化制合成气的研究-氧化铁的氧化还原性能[J]. 分子催化, 2001, 3(15): 181-186. (LI Ran-jia, SHENG Shi-kong. Study on lattice oxygen used in the conversion of methane to synthesis gas-redox performance of Fe₂O₃ catalyst[J]. Journal of Molecular Catalysis(China), 2001, 3(15): 181-186.)
- [13] NAKAYAMA O, IKENAGA N, MIYAKE T, YAGASAKI E, SUZUKI T. Production of synthesis gas from methane using lattice oxygen of NiO-Cr₂O₃-MgO complex oxide[J]. Ind Eng Chem Res, 2010, (492): 526-534.
- [14] 王华, 魏永刚. 晶格氧部分氧化甲烷制取合成气技术[M]. 北京: 冶金工业出版社, 2009: 88. (WANG Hua, WEI Yong-gang. Partial oxidation of methane by lattice oxygen to produce synthesis gas[M]. Beijing: Metallurgical Industry Press, 2009.)
- [15] 苏亚欣, 邓文义, 苏阿龙. 甲烷在氧化铁表面还原NO的特性与反应机理研究[J]. 燃料化学学报, 2013, 41(9): 1129-1135. (SU Ya-xin, DENG Wen-yi, SU A long. NO reduction by methane over iron oxides and the mechanism. Journal of Fuel Chemistry and Technology, 2013, 41(9): 1129-1135.)
- [16] SMOOT L D, HILL S C, XU H. NO_x control through reburning[J]. Prog Energy Combust Sci, 1998, 24(5): 385-408. 
- [17] 苏亚欣, GATHITU B B, CHEN W Y. Fe₂O₃控制再燃脱硝中间产物HCN的实验研究[J]. 环境科学学报, 2011, 31(6): 1181-1186. (SU Ya-xin, GATHITU B B, CHEN W Y. Experimental examination of HCN compound control by Fe₂O₃ during reburning processes[J]. Acta Scientiae Circumstantiae, 2011, 31(6): 1181-1186.)
- [18] TAN H Z, WANG X B, NIU Y Q, LIU H Y, WANG C L, XU T M. Studies of interreaction mechanism between iron and HCN[J]. Asian J Chem, 2010, 22(5): 4017-4025.
- [19] 陈庚. 气基还原氧化铁动力学机理研究[D]. 大连: 大连理工大学, 2011. (CHENG Geng. The kinetics of the gas-based reduction of iron oxide[D]. Dalian: Dalian University of Technology, 2011.)

[1] 刘自松, 魏永刚, 李孔斋, 王华, 祝星, 杜云鹏. Fe₂O₃/Al₂O₃氧载体用于甲烷化学链燃烧: 负载量与制备方法的影响[J]. 燃料化学学报, 2013, 41(11): 1384-1392.

[2] 王长真, 司兰杰, 李海, 闻霞, 孙楠楠, 赵宁, 魏伟, 孙予罕. 无模板剂一锅法合成介孔Ni-CaO-ZrO₂催化剂及其在CH₄-CO₂重整反应中的应用研究[J]. 燃料化学学报, 2013, 41(10): 1204-1209.

[3] 苏亚欣, 邓文义, 苏阿龙. 甲烷在氧化铁表面还原NO的特性与反应机理研究[J]. 燃料化学学报, 2013, 41(09): 1129-1135.

[4] 张俊峰, 白云星, 张清德, 解红娟, 谭猗生, 韩怡卓. Zr改性Ni/Y-Al₂O₃催化剂用于浆态相合成气的低温甲烷化[J]. 燃料化学学报, 2013, 41(08): 966-971.

- [5] 王永钊, 李凤梅, 程慧敏, 范莉渊, 赵永祥. 高镍负载量Ni/SiO₂和低镍负载量Ni-Ce/SiO₂催化CO甲烷化的比较研究[J]. 燃料化学学报, 2013, 41(08): 972-977.
- [6] 朱丽华, 徐锋, 王珏, 赵晓鹏. 醋酸溶液中Pd-CuPc/Y催化甲烷选择氧化制甲醇[J]. 燃料化学学报, 2013, 41(08): 985-990.
- [7] 陈宏刚, 王腾达, 张锴, 牛玉广, 杨勇平. 合成气甲烷化反应积炭过程的热力学分析[J]. 燃料化学学报, 2013, 41(08): 978-984.
- [8] 余钟亮, 李春玉, 景旭亮, 丁亮, 房倚天, 黄戒介. 碳酸钾催化的铁基氧载体煤催化化学链燃烧[J]. 燃料化学学报, 2013, 41(07): 826-831.
- [9] 余长林, 胡久彪, 杨凯, 周晓春. 制备方法对Ni/CeO₂-Al₂O₃催化剂甲烷部分氧化催化性能的影响[J]. 燃料化学学报, 2013, 41(06): 722-728.
- [10] 胡江亮, 孙天军, 任新宇, 常丽萍, 王树东. ZIF-8吸附剂上CH₄/N₂的吸附分离性能与热力学性质[J]. 燃料化学学报, 2013, 41(06): 754-760.
- [11] 熊志波, 郭东旭, 路春美, 张信莉. 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.
- [12] 毛菀钰, 孙启文, 应卫勇, 房鼎业. 高温沉淀铁基催化剂上费托合成含氧化合物生成机理的研究[J]. 燃料化学学报, 2013, 41(03): 314-322.
- [13] 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.
- [14] 由宏新, 高红杰, 陈刚, 阿布里提, 丁信伟. SOFC中干甲烷浓度对Ni-YSZ阳极上反应的影响[J]. 燃料化学学报, 2013, 41(03): 374-379.
- [15] 高帅, 郑青榕. 甲烷在活性炭上吸附平衡模型的研究[J]. 燃料化学学报, 2013, 41(03): 380-384.

版权所有 © 《燃料化学学报》编辑部

本系统由北京玛格泰克科技发展有限公司设计开发 技术支持: support@magtech.com.cn