

燃料化学学报 » 2014, Vol. 42 » Issue (03): 377-384 DOI:

研究论文

[最新目录](#) | [下期目录](#) | [过刊浏览](#) | [高级检索](#)

### SO<sub>2</sub>对甲烷在金属铁表面还原NO的反应影响

苏亚欣, 苏阿龙, 任立铭, 邓文义

东华大学 环境科学与工程学院, 上海 201620

### Effect of SO<sub>2</sub> on the reduction of NO by methane over iron catalyst

SU Ya-xin, SU A-long, REN Li-ming, 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](#) (814 KB) [HTML](#) (1 KB) 输出: [BibTeX](#) | [EndNote \(RIS\)](#) [背景资料](#)

**摘要** 采用卧式程序控温电加热陶瓷管反应器, 在N<sub>2</sub>和模拟烟气气氛中、300~1 100 ℃下, 研究了SO<sub>2</sub>对甲烷在金属铁及氧化物表面还原NO反应的影响。采用XRD等手段对反应前后铁催化剂样品的组成变化进行了表征, 分析了SO<sub>2</sub>在甲烷-铁脱硝中的作用机理。结果表明, 甲烷在金属铁及氧化铁表面能够高效率地还原NO, NO还原效率不受烟气中SO<sub>2</sub>的影响。在SO<sub>2</sub>分数为0.01%~0.04%的N<sub>2</sub>气氛中, 温度高于700 ℃时, 金属铁上NO还原率和SO<sub>2</sub>脱除率可同时达到100%。在SO<sub>2</sub>体积分数为0.01%~0.04%的模拟烟气中, 当温度高于850 ℃时, NO还原效率达到90%以上; 温度为950 ℃时, NO还原效率达到98%, SO<sub>2</sub>对NO还原效率的影响可忽略。当反应温度为1 000 ℃时, 在含0.02%SO<sub>2</sub>的模拟烟气中, 甲烷的体积分数为1时, 持续100 h金属铁表面上的NO还原效率都能保持95%以上。

**关键词:** NO还原 甲烷 铁 氧化铁 SO<sub>2</sub>

**Abstract:** The effect of SO<sub>2</sub> on the reduction of NO by methane over iron and iron oxides was investigated at 300~1 100 ℃ in an electrically heated ceramic tubular flow reactor in simulated flue gas and N<sub>2</sub> atmosphere. The iron catalyst after reaction was characterized by X-ray diffraction (XRD) and the mechanism of NO reduction by methane over iron catalyst in the presence of SO<sub>2</sub> was elucidated. The results demonstrated that methane is effective to reduce NO over iron and iron oxides and SO<sub>2</sub> in the flue gas has little influence on the reduction of NO. In N<sub>2</sub> atmosphere with 0.01%~0.04% SO<sub>2</sub>, metallic iron can simultaneously eliminate almost 100% of NO and SO<sub>2</sub> at a temperature above 700 ℃. In the simulated flue gas with 0.01%~0.04% SO<sub>2</sub>, the efficiencies of NO reduction at 850 ℃ and 950 ℃ are higher than 95% and 98%, respectively; the influence of SO<sub>2</sub> on NO reduction is insignificant. More than 95% of NO is reduced 1.13% methane over iron at 1 000 ℃ in a durable test over 100 h in the simulated flue gas atmosphere containing 0.02% SO<sub>2</sub>.

**Key words:** NO reduction methane iron iron oxide SO<sub>2</sub>

收稿日期: 2013-08-06;

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, 教授, E-mail: suyx@dhu.edu.cn, Tel: 021-67792552 E-mail: suyx@dhu.edu.cn

引用本文:

苏亚欣, 苏阿龙, 任立铭等. SO<sub>2</sub>对甲烷在金属铁表面还原NO的反应影响[J]. 燃料化学学报, 2014, 42(03): 377-384.

- [1] PARVULESCU V I, GRANGE P, DELMON B. Catalytic removal of NO[J]. *Catal Today*, 1998, 46(4): 233
- [2] JANSSEN F, MEIJER R. Quality control of DeNO<sub>x</sub> catalysts performance testing, surface analysis and [J]. *Catal Today*, 1993, 16(2): 157-185. 
- [3] IWAMOTO M. Zeolites in environmental catalysis[J]. *Stud Surf Sci Catal*, 1994, 84: 1395-1410. 
- [4] TABATA T, KOKITSU M, OKADA O. Study on patent literature of catalysts for a new NO<sub>x</sub> removal p 147-169. 
- [5] FENG X, HALL W K. FeZSM-5: A durable SCR catalyst for NO<sub>x</sub> removal from combustion streams[J]. 
- [6] APOSTOLESCU N, GEIGER B, HIZBULLAH K, JAN M. T, KURETI S, REICHERT D, SCHOTT F, WEISWE of nitrogen oxides by ammonia on iron oxide catalysts[J]. *Appl Catal B: Environ*, 2006, 62(1/2): 104
- [7] KRISHNA K, SEIJGER G B F, VAN DEN BLEEK C M, MAKKEE M, MUL G, CALIS H P A. Selective cataly ZSM-5 catalysts prepared by sublimation of FeCl<sub>3</sub> at different temperatures[J]. *Catal Lett*, 2003, 8
- [8] BETHKE K A, KUNG M C, YANG B, SHAH M, ALT D, LI C, KUNG H H. Metal oxide catalysts for lean NC 26(2): 169-183. 
- [9] BETHKE K A, ALT D, KUNG M C. NO reduction by hydrocarbons in an oxidizing atmosphere over tran [J]. *Catal Lett*, 1994, 25(1/2): 37-48. 
- [10] ILIOPOULOU E F, EVDOU A P, LEMONIDOU A A, VASALOS I A. Ag/alumina catalysts for the selectiv various reductants[J]. *Appl Catal A: Gen*, 2004, 274(1/2): 179-189. 
- [11] KOTSIFA A, KONDARIDES D I, VERYKIOS X E. A comparative study of the selective catalytic reduct supported Pt and Rh catalysts[J]. *Appl Catal B: Environ*, 2008, 80(3/4): 260-270. 
- [12] LIU Z, WANG K, ZHANG X, WANG J, CAO H, GONG M, CHEN Y. Study on methane selective catalyt Pt/Ce<sub>0.67</sub>Zr<sub>0.33</sub>O<sub>2</sub> and its application[J]. *J Nat Gas Chem*, 2009, 18(1): 66-70. 
- [13] SMOOT L D, HILL S C, XU H. NO<sub>x</sub> control through reburning[J]. *Prog Energ Combus Sci*, 1998, 24(5)
- [14] 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): 112' A-long. NO reduction by methane over iron oxides and the mechanism[J]. *Journal of Fuel Chemistry* 1135.)
- [15] 苏亚欣, 任立铭, 苏阿龙, 邓文义. 甲烷在金属铁及氧化铁表面还原NO的实验研究[J]. 燃料化学学报, 2013, 41(11 ming, SU A-long, DENG Wen-yi. Experimental study on NO reduction by methane over iron and its c and Technology, 2013, 41(11): 1393-1400.) 浏览
- [16] BIRKS N, MEIER G H. Introduction to high temperature oxidation of meta[M]. Edward Arnold, London
- [17] FLYTZANI-STEFANOPoulos M, ZHU T, LI Y. Ceria-based catalysts for the recovery of elemental : [J]. *Catal Today*, 2000, 62(2/3): 145-158. 
- [18] ZHU T, DREHER A, FLYTZANI-STEFANOPoulos M. Direct reduction of SO<sub>2</sub> to elemental sulfur by [J]. *Appl Catal B: Environ*, 1999, 21(2): 103-120. 
- [19] ZHU T, KUNDAKOVIC L, DREHER A, FLYTZANI-STEFANOPoulos M. Redox chemistry over CeO<sub>2</sub>-ba or CH<sub>4</sub>[J]. *Catal Today*, 1999, 50(2): 381-397. 
- [20] SARLIS J, BERK D. Reduction of sulfur dioxide by methane over transition metal oxide catalysts[J]. C 73-85.
- [21] 邓庚凤, 姜坤, 曹霞, 肖文仲, 何桂荣. 煤气还原冶炼烟气中SO<sub>2</sub>制取硫磺的工艺研究[J]. 江西理工大学学报, 2010, JIANG Kun, CAO Xia, XIAO Wen-zhong, HE Gui-rong. Study on the catalytic reduction of SO<sub>2</sub> in the Journal of Jiangxi University of Science and Technology, 2010, 31(1): 32-35.)
- [22] 胡大为, 秦永宁, 马智, 齐晓周, 张黎明. 负载型过渡金属氧化物催化剂上SO<sub>2</sub>的还原[J]. 燃料化学学报, 2001, 29 ning, MA Zhi, QI Xiao-zhou, ZHANG Li-ming. Reduction of sulfur dioxide over supported transition m Chemistry and Technology, 2001, 29(6): 499-503.)
- [23] 胡大为, 秦永宁, 马智, 韩森. 载体对CO还原SO<sub>2</sub>到单质硫铁基催化剂性能的影响[J]. 燃料化学学报, 2002, 30(2) ning, MA Zhi, HAN Sen. Effect of support on catalytic property of Fe catalyst for sulfur dioxide redi monoxide[J]. *Journal of Fuel Chemistry and Technology*, 2002, 30(2): 156-161.)
- [24] PAN Y G, PERALES J F, VELO E, PUIGJANER L. Kinetic behaviour of iron oxide sorbent in hot gas des 1105-1109. 

- [25] TAMHANKAR S S, HASATANI M, WEN C Y. Kinetic-studies on the reactions involved in the hot gas c iron-oxide sorbent.1. Reduction and sulfidation of iron-oxide[J]. Chem Eng Sci, 1981, 36(7): 1181-'
- [26] 黄吉庆, 白宗庆, 白进, 郭振兴, 李文. 过渡金属添加剂对煤热解脱硫的影响[J]. 燃料化学学报, 2012, 40(6): 641  
BAI Jin, GUO Zhen-xing, LI Wen. Effects of transition metal additives on desulfurization of coal in p and Technology, 2012, 40(6): 641-647.)
- [27] CHEAH S, CARPENTER D L, MAGRINI-BAIR K A. Review of mid-to high-temperature sulfur sorbents for coal-derived syngas[J]. Energy Fuels, 2009, 23(11): 5291-5307. 

[1] 孟凡会, 刘军, 李忠, 钟朋展, 郑华艳. Ce含量对Ni-Ce/Al<sub>2</sub>O<sub>3</sub>催化剂结构及浆态床CO甲烷化性能的影响[J].  
237.

[2] 张军伟, 黄戒介, 房倚天, 王志青, 余钟亮. 镧修饰铁基复合载氧体用于化学链甲烷部分氧化重整制合成气研究  
165.

铁酸镍用于热化学循环反应CO 分解制CO的研究