

ZSM-5的水热改性及其在合成气经二甲醚制汽油中的应用

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Hydrothermal treatment of ZSM-5 and its application in syngas to gasoline via DME

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摘要 对合成气经二甲醚制汽油两段等压反应系统中的ZSM-5分子筛催化剂进行原位水热处理,研究了不同温度下水蒸气处理ZSM-5分子筛的结构、表面酸性及催化反应性能。XRD、XRF、BET、NH₃-TPD 和 TPO等表征手段表明,水热处理温度为400℃时会使ZSM-5分子筛大部分非骨架铝脱除,酸中心数量减少,比表面积增加,提高了催化剂的活性及产物选择性。

关键词: ZSM-5 水热处理 合成气 二甲醚 汽油

Abstract: The effects of in-situ hydrothermal treatment on the ZSM-5 zeolite were studied, and its performance for syngas to gasoline via DME reaction process was discussed using two-stage equal-pressure reactor system. The texture, surface acidity and catalytic performance of the treated ZSM-5 zeolite at different temperatures were investigated. Physicochemical properties of the zeolite were characterized by XRD, XRF, BET, NH₃-TPD and TPO. The results indicated that most of Al was removed from the nonframework of zeolite through appropriate hydrothermal treatment at 400℃. A decrease in the amount of acidic sites and an increase of specific surface area were observed, and accordingly, the catalytic activity of the ZSM-5 zeolite and product selectivity were improved significantly.

Key words: ZSM-5 hydrothermal treatment syngas DME gasoline

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[1] KUNG H H. Methanol synthesis[J]. Cat Rev-Sci Eng, 1980, 22(2): 235-259. 

[2] KANG S H, BAE J W, JUN K W, POTDAR H S. Dimethyl ether synthesis from syngas over the composite catalysts of Cu-ZnO-Al₂O₃/Zr-

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modified zeolites[J]. Catal Commun, 2008, 9(10): 2035-2039. 

- [3] ANDRÉS G T, AGUSTÍN M. Direct synthesis of DME from syngas on hybrid CuZnAl/ZSM-5 catalysts: New insights into the role of zeolite acidity[J]. Appl Catal A: Gen, 2012, 411-412: 170-179. 
- [4] 张清德, 谭猗生, 杨彩虹, 韩怡卓. 反应压力对HZSM-5分子筛催化剂上合成气经二甲醚制取芳烃反应的影响[J]. 现代化工, 2009, 29(S1): 112-114. (ZHANG Qing-de, TAN Yi-sheng, YANG Cai-hong, HAN Yi-zhuo. Effect of reaction pressure for syngas to aromatics via dimethyl ether over HZSM-5 catalyst[J]. Modern Chemical Industry, 2009, 29(S1): 112-114.)
- [5] KOLESNICHENKO N V, KITAEV L E, BUKINA Z M, MARKOVA N A, YUSHCHENKO V V, YASHINA O V, LIN G I, ROZOVSKII A Y. Synthesis of gasoline from syngas via dimethyl ether[J]. Kinet Catal, 2007, 48(6): 789-793. 
- [6] FUJIMOTO K, ASAMI K, SAIMA H, SHIKADA T, TOMINAGA H. Two-stage reaction system for synthesis gas conversion to gasoline[J]. Ind Eng Chem Pro Res Dev, 1986, 25(2): 262-267. 
- [7] 岳瑛, 丁莹如. 用改良MTG方法自合成气两步合成汽油催化剂的研究[J]. 燃料化学学报, 1990, 18(3): 247-252. (YUE Ying, DING Ying-ru. Study on catalysts for two stage gasoline synthesis from syngas[J]. Journal of Fuel Chemistry and Technology, 1990, 18(3): 247-252.) 
- [8] TOSHEVA L, VALTCHEV V P. Nanozeolites: Synthesis, crystallization mechanism, and applications[J]. Chem Mat, 2005, 17(10): 2494-2513. 
- [9] SONG W, JUSTICE R E, JONES C A, GRASSIAN V H, LARSEN S C. Synthesis, characterization, and adsorption properties of nanocrystalline ZSM-5[J]. Langmuir, 2004, 20(19): 8301-8306. 
- [10] LARSEN S C. Nanocrystalline zeolites and zeolite structures: Synthesis, characterization, and applications[J]. J Phys Chem C, 2007, 111(50): 18464-18474. 
- [11] 盛清涛, 凌开成, 李振荣, 赵亮富. 水蒸汽及盐酸处理对ZSM-5分子筛性能的影响[J]. 太原理工大学学报, 2012, 43(4): 425-430. (SHENG Qing-tao, LING Kai-cheng, LI Zhen-rong, ZHAO Liang-fu. Effect of steam treatment and HCl solution treatment on catalytic performance of ZSM-5 zeolite for the conversion of ethanol to ethylene[J]. Journal of Tai Yuan University of Technology, 2012, 43(4): 425-430.)
- [12] 张培青, 王祥生, 郭洪臣, 徐舟波, 赵乐平, 胡永康. 水热处理对纳米HZSM-5沸石酸性质及其降低汽油烯烃性能的影响[J]. 催化学报, 2003, 24(12): 900-904. (ZHANG Pei-qing, WANG Xiang-sheng, GUO Hong-chen, XU Zhou-bo, ZHAO Le-ping, HU Yong-kang. Influence of hydrothermal treatment on acid property and catalytic performance of nano-HZSM-5 zeolite for reducing olefins in gasoline[J]. Chinese Journal of Catalysis, 2003, 24(12): 900-904.) 
- [13] 汪树军, 梁娟, 郭文珪, 赵素琴, 汪荣慧. ZSM-5沸石骨架铝迁移规律的研究: I.水热处理条件及沸石硅铝比的影响[J]. 催化学报, 1992, 13(1): 38-43. (WANG Shu-jun, LIANG Juan, GUO Wen-gui, ZHAO Su-qin, WANG Rong-hui. Studies on the migration of framework Al of ZSM-5 zeolite: I. Effect of hydrothermal treatment and silica alumina ratio[J]. Chinese Journal of Catalysis, 1992, 13(1): 38-43.)
- [14] DING C H, WANG X S, GUO X W, ZHANG S G. Characterization and catalytic alkylation of hydrothermally dealuminated nanoscale ZSM-5 zeolite catalyst[J]. Catal Commun, 2008, 9(4): 487-493. 
- [15] DATKA J, MARSCHMEYER S, NEUBAUER T, MEUSINGER J, PAPP H, SCHUTZE F W, SZPYT I. Physicochemical and catalytic properties of HZSM-5 zeolites dealuminated by the treatment with steam[J]. J Phys Chem, 1996, 100(34): 14451-14456. 
- [16] 李剑, 谭猗生, 杨彩虹, 韩怡卓. 水对两段法合成汽油中HZSM-5分子筛稳定性及油相产物的影响[J]. 燃料化学学报, 2010, 38(1): 96-101. (LI Jian, TAN Yi-sheng, YANG Cai-hong, HAN Yi-zhuo. Effect of water on stability of HZSM-5 and product distribution in two-stage gasoline synthesis from syngas [J]. Journal of Fuel Chemistry and Technology, 2010, 38(1): 96-101.)
- [17] ONG L H, DOMOK M, OLINDO R, VAN VEEN A C, LERCHER J A. Dealumination of HZSM-5 via steam-treatment[J]. Micropor Mesopor mater, 2012, 164: 9-20. 
- [18] EROFEEV V I, ADYAEVA L V, RYABOVA N V. Effect of high-temperature steam treatment of high-silica zeolites of the ZSM-5 type on their acidity and selectivity of formation of lower olefins from straight-run naphthas[J]. Russ J Appl Chem, 2003, 76(1): 95-98. 
- [19] 杨成, 李建青, 李琢, 李超, 孙振鹏, 吴晋沪, 王利生. 一种用于合成气制二甲醚的催化剂及其制法和应用: 中国, 101940934A[P]. 2011-01-02. (YANG Cheng, LI Jian-qing, LI Zhuo, LI Chao, SUN Zhen-peng, WU Jin-hu, WANG Li-sheng. A kind of catalyst used for the synthesis gas to dimethyl ether and its preparation and application: CN, 101940934A[P]. 2011-01-02.)
- [1] 赵晓波, 王文举, 郭新闻, 王祥生. Al_2O_3 孔结构对纳米HZSM-5基催化剂改质FCC汽油性能的影响[J]. 燃料化学学报, 2013, 41(11): 1343-1348.
- [2] 张君涛, 郝娜娜, 王妮. ZSM-5/MCM-41复合分子筛的合成与表征[J]. 燃料化学学报, 2013, 41(10): 1268-1273.
- [3] 海航, 闫常峰, 胡蓉蓉, 郭常青, 李文博. 助剂对泡沫金属微反应器内二甲醚水蒸气重整制氢的影响[J]. 燃料化学学报, 2013, 41(10): 1210-1216.
- [4] 赵岑, 刘冬梅, 魏民, 孙志岩, 王海彦. 多级孔ZSM-5分子筛的制备及催化噻吩烷基化性能研究[J]. 燃料化学学报, 2013, 41(10): 1256-1261.
- [5] 孙明, 余林, 那秀辉, 刁贵强, 叶飞. OMS-2/ Al_2O_3 /堇青石整体式催化剂的制备及其催化性能研究[J]. 燃料化学学报, 2013, 41(09): 1115-1121.
- [6] 王文丽, 王琰, 陈月仙, 赵文超, 李瑞丰. 合成气一步法制备二甲醚核壳结构催化剂的制备及其反应性能[J]. 燃料化学学报, 2013, 41(08): 1003-1009.
- [7] 马现刚, 葛庆杰, 徐恒泳. 复合催化剂上合成气一步法制备液化石油气的研究[J]. 燃料化学学报, 2013, 41(08): 1010-1014.
- [8] 张俊峰, 白云星, 张清德, 解红娟, 谭猗生, 韩怡卓. Zr改性Ni/ γ - Al_2O_3 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.
- [9] 武应全, 解红娟, 寇永利, 谭理, 韩怡卓, 谭猗生. 焙烧温度对K-Cu/Zn/La/ ZrO_2 催化剂上异丁醇合成的影响[J]. 燃料化学学报, 2013, 41(07): 868-874.
- [10] 王琰, 王文丽, 陈月仙, 郑家军, 李瑞丰. 含有多级孔复合分子筛的复合催化剂上合成气一步制二甲醚[J]. 燃料化学学报, 2013, 41(07): 875-882.

- [11] 寇永利, 解红娟, 刘广波, 武应全, 张欣悦, 韩怡卓, Noritatsu Tsubaki, 谭猗生. ZnCr基催化剂煅烧温度对异丁醇合成性能的影响[J]. 燃料化学学报, 2013, 41(06): 703-709.
- [12] 石冈, 林秀英, 范煜, 鲍晓军. ZSM-5分子筛的脱硅改性及加氢改质性能[J]. 燃料化学学报, 2013, 41(05): 589-600.
- [13] 杨瑞娟, 杨冬花, 武正簧, 冯晓娜, 窦涛, 吴忠华, 李志宏, 闫泽, 王凡, 汤琼. 含铁微孔EU-1/ZSM-5复合分子筛的合成和表征[J]. 燃料化学学报, 2013, 41(05): 601-606.
- [14] 傲云宝勒德, 张楹斗, 周晨亮, 李阳, 陈琛, 智科端, 宋银敏, 滕英跃, 何润霞, 刘全生. 蒙古国巴嘎诺尔(Baganuur)褐煤水蒸气气化制富氢合成气及其固有矿物质的催化作用[J]. 燃料化学学报, 2013, 41(04): 414-421.
- [15] 赵强, 李为民, 陈清林. Brønsted酸性离子液体催化合成聚甲醛二甲醚的研究[J]. 燃料化学学报, 2013, 41(04): 463-468.

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