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介孔硅胶在柴油氧化-吸附组合脱硫中的应用研究

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Application of mesoporous silica gel in desulfurization of diesel oil via oxidation-adsorption process

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摘要 以硅胶(SG)为吸附剂,采用自制的双亲催化剂与H_2O_2组成的催化氧化体系将柴油进行氧化,利用固定床动态吸附法考察了硅胶性质、氧化过程及吸附条件等对硅胶吸附脱硫性能的影响,并对硅胶进行了表征。小角XRD和氮气吸脱附结果表明,实验所用硅胶具有介孔结构。吸附脱硫实验结果表明,在油剂比(柴油与吸附剂的体积比)相同时,氧化-吸附脱硫过程脱硫率明显高于吸附脱硫过程脱硫率;选用硅胶作吸附剂,吸附温度为40℃,吸附空速为6.0 h^{-1}时脱硫效果较好,当油剂比为1时,脱硫率高达94.57%,且该介孔硅胶具有较大的吸附硫容,随油剂比增大下降缓慢,当油剂比增大到15时,脱硫率仍达85.89%。

关键词: 介孔硅胶 氧化-吸附 脱硫 柴油

Abstract: The desulfurization of diesel oil was conducted via oxidation-adsorption process, using H_2O_2 as oxidant in the presence of amphiphilic catalyst and mesoporous silica-gel as the adsorbent. The fixed-bed dynamic adsorption method was employed to evaluate the oxidation-adsorption desulfurization capability of the catalysts and the effects of the property of silica-gel, oxidation and adsorption conditions on desulfurization efficiency were investigated. It was proved that the silica-gel possessed the mesoporous structure characterized by small-angle XRD and N_2 adsorption-desorption isotherm. Compared with adsorption process, oxidation-adsorption process could availably increase the adsorptive capacity and selectivity of silica-gel adsorbent to sulfur compounds in diesel oil.The sulfur removal efficiency remained as high as 85.89% when the volume ratio of diesel oil to silica-gel was 15. Sulfur removal efficiency reached up to 94.57% when adsorption temperature, space velocity and volume ratio of diesel oil to silica-gel were 40℃, 6.0 h^{-1} and 1 respectively.

Key words: mesoporous silica gel oxidation-adsorption desulfurization diesel oil

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[1] 郝临山, 彭建喜. 洁净煤技术[M]. 北京: 化学工业出版社, 2010. (HAO Lin-shan, PENG Jian-xi. Clean coal technology[M]. Beijing: Chemical Industry Press, 2010.)

[2] 李开喜, 吕春祥, 凌立成. 活性炭纤维的脱硫性能[J]. 燃料化学学报, 2002, 30(1): 89-96. (LI Kai-xi, LV Chun-xiang, LING Li-cheng. Activity of activated carbon fiber for SO_2 removal[J]. Journal of Fuel Chemistry and Technology, 2002, 30(1): 89-96.)

[3] MOCHIDA I, KAWABUCHI Y, KAWANO S. High catalytic activity of pitch-based activated carbon fibres of moderate surface area for oxidation of NO to NO_2 at room temperature[J]. Fuel, 1997, 76(6): 543-548.

[4] 张守臣, 赵修仁, 刘长厚, 郭世永, 陈家骅. 硝酸浸渍对活性炭纤维还原NO性能的影响[J]. 化工学报, 2001, 52(8): 690-695. (ZHANG Shou-chen, ZHAO Xiu-ren, LIU Chang-hou, GUO Shi-yong,CHEN Jia-hua. Effect of HNO_3 dipping on NO reduction properties over ACF[J]. Journal of Chemical Industry and Engineering(China), 2001, 52(8): 690-695.)

[5] SONG C. An overview of new approaches to deep desulfurization for ultra-clean gasoline, diesel fuel and jet fuel[J]. Catalysis Today, 2003, 86(1/4): 211-263.

[6] PARK S J, KIM B J. Role of acidic function groups of carbon fiber surfaces in enhancing interfacial adhesion behavior[J]. Mater Sci Eng, 2005, 408(1): 269-273.

[7] 王少军, 凌凤香, 王安杰. 柴油非加氢脱硫技术研究中样品的选择[J], 燃料化学学报, 2005, 33(2): 171-174. (WANG Shao-jun, LING Feng-xiang, WANG An-jie. Testing sample choice for non-hydrodesulfurization of diesels[J]. Journal of Fuel Chemistry and Technology, 2005, 33(2): 171-174.)

[8] DELIYANNI E, BANDOSZ T J. Effect of carbon surface modification with dimethyl lamine on reactive adsorption of NO_x[J]. Langmuir, 2011, 27(5): 1837-1843.

[9] 沈伯雄, 周元驰, 史展亮, 杨婷婷. 活性炭纤维的预处理及其SCR催化活性研究[J]. 燃料化学学报, 2008, 36(3): 376-380. (SHEN Bo-xiong, ZHOU Yuan-chi, SHI Zhan-liang, YANG Ting-ting. Study on the pretreatment of activated carbon fibers and its activity in SCR[J]. Journal of Fuel Chemistry and Technology, 2008, 36(3): 376-380.)

[10] YAZU K, FURUYA T, MIKI K, UKRGAWAK. Tungstophosphoric acid catalyzed oxidative desulfurization of light oil with hydrogen peroxide in a ligh t oil/ acetic acid biphasic system[J]. Chem Lett, 2003, 32(10): 920-921.

[11] 查庆芳, 高南星, 李兆丰, 卓海波, 张玉贞. 石油焦系活性炭的吸附脱硫[J]. 燃料化学学报, 2007, 35(2): 192-197. (ZHA Qing-fang, GAO Nan-xing, LI Zhao-feng, ZHUO Hai-bo, ZHANG Yu-zhen. Adsorption desulfurization by activated carbon from petroleum coke[J]. Journal of Fuel Chemistry and Technology, 2007, 35(2): 192-197.)

[12] ADAPA S, GAUR V, VERMA N. Catalytic oxidation of NO by activated carbon fiber(ACF)[J]. Chem Eng J, 2006, 116(1): 25-37.

[13] 王建龙, 赵地顺, 周二鹏, 董芝. 吡啶类离子液体在汽油萃取脱硫中的应用研究[J]. 燃料化学学报, 2007, 35(3): 293-296. (WANG Jian-long, ZHAO Di-shun, ZHOU Er-peng, DONG Zhi. Desulfurization of gasoline by extraction with N-alkyl-pyridinium-based ionic liquids[J]. Journal of Fuel Chemistry and Technology, 2007, 35(3): 293-296.)

[14] 陈永. 多孔材料制备与表征[M]. 合肥: 中国科学技术大学出版社, 2010. (CHEN Yong. Preparation and characterization of porous materials[M]. Hefei: University of Science & Technology of China Press, 2010.)

[15] 刘振宇, 郑经堂, 王茂章. 多孔炭的纳米结构及其解析[J]. 化学进展, 2001, 13(1): 10-18. (LIU Zhen-yu, ZHENG Jing-tang, WANG Mao-zhang. Nanostructure and analysis of porous carbons[J].Progress in Chemistry, 2001, 13(1): 10-18.)

[16] 吴永文, 李忠, 奚红霞, 李祥斌, 韩静磊, 郭建光. 高聚物吸附剂的孔隙结构和表面特性对苯酚吸附容量的影响[J]. 化工学报, 2003, 54(11): 1642-1645. (WU Yong-wen, LI Zhong, XI Hong-xia, LI Xiang-bin, HAN Jing-lei, GUO Jian-guang. Effect of porosity and surface properties of polymeric resins on adsorption capacity of phenol[J]. Journal of Chemical Industry and Engineering(China), 2003, 54(11): 1642-1645.)

[17] 杨全红, 郑经堂, 王茂章, 张碧江. 二次炭化对PAN-ACF结构和性能的影响[J]. 离子交换与吸附, 1999, 15(5): 385-390. (YANG Quan-hong, ZHENG Jing-tang, WANG Mao-zhang, ZHANG Bi-jiang. Influence of post-carbonization on structures and properties of ACFs[J]. Ion Exchange and Adsorption, 1999, 15(5): 385-390.)

[18] 黄艳芳, 马正飞, 姚虎卿. 活性炭吸附CO_2与其微孔体积的关系[J]. 燃料化学学报, 2008, 36(3): 343-348. (HUANG Yan-fang, MA Zheng-fei, YAO Hu-qing. Relation between adsorption of CO_2 on activated carbon and its micropore volume[J], 2008, 36(3): 343-348.)

[19] 任建莉, 陈俊杰, 罗誉娅, 何胜, 钟英杰. 活性炭纤维脱除烟气中气态汞的试验研究[J]. 中国电机工程学报, 2010, 30(5): 28-34. (REN Jian-li, CHEN Jun-jie, LUO Yu-ya, HE Sheng, ZHONG Ying-jie. An experimental study on activated carbon sorbents for gas-phase mercury removal from flue gas[J]. Proceedings of the CSEE, 2004, 24(2): 171-175.)

[20] ISHIHARA A, WANG D, DUMEIGNIL F, AMANO H, QIAN EW, KABE T. Oxidative desulfurization and denitrogenation of a light gas oil using an oxidation/adsorption continuous flow process[J]. Appl Catal A, 2005, 279(1/2): 279-287.

[21] RAYMUNDO-PINERO E, CAZORLA-AMOROS D, LINARES-SOLANO A. The role of different nitrogen functional groups on the removal of SO_2 from flue gases by N-doped activated carbon powders and fibres[J]. Carbon, 2003, 41(10): 1925-1932.

[22] MA X, VELU S, KIM J, SONG C. Deep desulfurization of gasoline by selective adsorption over solid adsorbents and impact of analytical methods on ppm-level sulfur quantification for fuel cell applications [J]. Appl Catal B, 2005, 56(1/2): 137-147.

[23] 冯辉, 曾勇平, 居沈贵. 载铜5A分子筛在汽油模拟体系中脱硫性能的研究[J]. 燃料化学学报, 2006, 34(1): 117-119. (FENG Hui, ZENG Yong-ping, JU Shen-gui. Desulfurization of model gasoline by 5A molecular sieves loaded with Cu^{2+} [J]. Journal of Fuel Chemistry and Technology, 2006, 34(1): 117-119.)

[24] 孟启, 孙小强, 姜艳, 席海涛, 杨绪杰, 陆路德, 汪信. 磺酸型树脂酰胺化修饰及修饰产物的吸附脱硫研究[J]. 燃料化学学报, 2007, 35(2): 249-252. (MENG Qi, SUN Xiao-qiang, JIANG Yan, XI Hai-tao, YANG Xu-jie, LU Lu-de, WANG Xin. Adsorptive desulfurization properties of sulfuric resin with amidat modification[J]. Journal of Fuel Chemistry and Technology, 2007, 35(2): 249-252.)

[25] 何余生, 李忠, 奚红霞, 郭建光, 夏启斌. 气固吸附等温线的研究进展[J]. 离子交换与吸附, 2004, 20(4):376-384. (He Yu-sheng, Li Zhong, Xi Hong-xia, Guo Jian-guang, Xi Qi-bin. Research progress of gas-solid adsorption isotherms[J]. Ion Exchange and Adsorption, 2004, 20(4): 376-384.)

[26] MOCHIDA I, KISAMORI S, HIRONAKA M. Oxidation of NO into NO_2 over active carbon fibers[J].Energy Fuels, 1994, 8(3): 1341-1344.

[27] 汪政德, 张茂林, 梅海燕. 毛细凝聚和吸附-脱附回路的物理化学解释[J]. 新疆石油地质, 2002, 23(3): 233-236. (WANG ZHENG-de, ZHANG Mao-lin, MEI Hai-yan. The physical chemistry explanation of the capillary condensation and the circuit of adsorption-desorption[J]. Xinjiang Petroleum Geology, 2002, 23(3): 233-236.)

[28] GARCIA P, ESPINAL J F, SALINAS C, de LECEA M, MONDRAGON E. Experimental characterization and molecular simulation of nitrogen complexes formed upon NO-char reaction at 270 ℃ in the presence of H_2O and O_2[J]. Carbon, 2004, 42(8): 1507-1515.

[29] GROEN J C, P REZ-RAMIREZ. Critical Appraisal of mesopore characterization by adsorption analysis[J]. Appl Catal A, 2004, 268(1/2): 121-125.

[30] 王幸宜. 催化剂表征[M]. 上海: 华东理工大学出版社, 2008. (WANG Xing-yi. Characterization of the catalyst[M]. Shanghai: East China University of Science and Technology Press, 2008.)

[31] 宁永成. 有机化合物结构鉴定与有机波谱学[M]. 2版. 北京: 科学出版社, 2008: 322-363. (NING Yong-cheng. Structural identification of organic compounds and organic spectroscopy[M]. 2nd ed. Beijing: Science Press, 2008: 322-363.)

[1] 左华亮, 刘琪英, 王铁军, 史娜, 刘建国, 马隆龙. 负载的Ni催化剂上植物油脂加氢脱氧制备第二代生物柴油[J]. 燃料化学学报, 2012, 40(09): 1067-1073.

[2] 郭姣姣, 杨永珍, 张永奇, 房倚天, 刘旭光. 正交实验研究硝酸改性对活性焦表面性质及脱硫性能的影响[J]. 燃料化学学报, 2012, (08): 1014-1018.

[3] 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(06): 641-647.

[4] 武宝萍, 沈健, 张秋荣. 微孔-介孔复合分子筛HY-SBA-15的表征及应用[J]. 燃料化学学报, 2012, 40(06): 732-736.

[5] 周亚松, 张涛, 魏强. 络合作用对重油加氢处理催化剂性能的影响[J]. 燃料化学学报, 2012, 40(05): 621-625.

[6] 张薇, 丁永萍, 宫敬, 宋溪明. 羧基功能化离子液体催化二苯并噻吩氧化脱硫[J]. 燃料化学学报, 2012, 40(05): 626-629.

[7] 王利兵, 于海燕, 贺晓辉, 刘瑞英. 生物柴油树种油脂脂肪酸组成对燃料特性的影响[J]. 燃料化学学报, 2012, (04): 397-404.

[8] 李梅, 吕鹏梅, 杨玲梅, 罗文, 王忠铭, 李惠文, 袁振宏. Fe-Ca磁性固体碱催化剂上菜籽油酯交换反应[J]. 燃料化学学报, 2012, (04): 405-412.

[9] 陈颖, 孙雪, 李慧, 白云波. 稀土改性对SO_4^{2-}/ZrO_2固体酸催化剂结构与催化性能的影响[J]. 燃料化学学报, 2012, (04): 412-417.

[10] 李翠清, 邓明亮, 王虹, 葛明兰, 郭志武, 孙桂大. 含磷铝单元的AlMCM-41负载的磷化钨催化剂DBT HDS性能[J]. 燃料化学学报, 2012, (04): 501-506.

[11] 唐克, 洪新, 宋丽娟, 孙兆林. 二次合成Y型杂原子分子筛的吸附脱硫研究[J]. 燃料化学学报, 2012, (04): 507-512.

[12] 孟璇, SALISSOU ZAKATY M N, 翁惠新, 施力. 不同载体的镍基氧化锌对噻吩的吸附脱除性能[J]. 燃料化学学报, 2012, 40(03): 364-369.

[13] 郑华艳, 李茜茜, 崔丽萍, 李忠. Ca/Al固体碱催化菜籽油制备生物柴油[J]. 燃料化学学报, 2012, 40(03): 331-336.

[14] 朱健鹏, 李春虎, 陈佳玲, 罗莹玮, 冯丽娟, 孔玉普. 常温常压D072树脂氧化吸附油品脱硫的研究[J]. 燃料化学学报, 2012, 40(03): 370-373.

[15] 赵策, 曾虹燕, 黄炎, 刘平乐, 王亚举, 杨永杰, 张伟. 镁铁水滑石的制备及其对小球藻油脂合成生物柴油的催化性能[J]. 燃料化学学报, 2012, 40(03): 337-344.