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论文

# CuHY分子筛中铜离子的分布与吸附脱硫性能

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摘要:

采用等体积浸渍法制备具有不同Cu担载量的CuHY 分子筛吸附剂. 用X射线衍射(XRD)、比表面积(BET)和氨程序升温脱附($NH_3$-TPD)技术对分子筛吸附剂进行了表征, 并测定了CuHY 分子筛吸附剂在含二苯并噻吩(DBT)模拟柴油中的吸附脱硫性能; 通过多晶XRD确定了$Cu^{2+}$在Cu8HY 分子筛笼内的结构与分布. 实验结果表明, 分子筛的骨架结构没有发生改变, 部分$Cu^{2+}$进入Y型分子筛笼内, 分子筛样品强酸中心有所减少, 中强酸中心有所增加; 进入Y型分子筛笼内的Cu2+, 一部分处于β 笼的$S_I$, 位, 另一部分位于分子筛超笼中的$S_{III}$位上, 并与笼内的水分子配位. 处于超笼中的$S_{III}$位$Cu^{2+}$对模拟柴油中的DBT分子具有吸附作用, 是吸附脱硫的中心. 而当模拟柴油中存在萘时, 与DBT分子会产生竞争吸附.

关键词: CuHY分子筛吸附剂 二苯并噻吩(DBT) 吸附脱硫

# Distribution of the Cu Ions in the CuHY Zeolite and Its Performance of Desulfurization

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Abstract:

The CuHY zeolites' absorbents with different Cu loadings were prepared *via* isovolumic impregnation method, and characterized *via* X-ray diffraction(XRD), BET surface area measurement and $NH_3$ temperature-programmed desorption($NH_3$-TPD) techniques. The CuHY zeolites absorbents' performance of desulfurization was investigated in the model diseles containing dibenzothiophene(DBT). The crystalline structure of Cu8HY zeolite and the $Cu^{2+}$ cations distribution in the cages of the Y zeolite was determined *via* powder XRD with Rietveld method. The results show that the frameworks of the Y zeolites were retained after $Cu^{2+}$ cations entered the cages of the Y zeolites, the amounts of strong acid sites increased slightly and the amounts of mid-strong acid sites decreased slightly in the CuHY zeolite. At the same time, one part of $Cu^{2+}$ cations entering the cages of Y zeolite, and situated at the sites $S_I$, in the *β* cages, while another part of $Cu^{2+}$ cations situated at sites $S_{III}$ in the supercages and coordinated with water molecules in the supercages. The $Cu^{2+}$ cations situating at sites $S_{III}$ in the supercages can adsorb DBT molecules in the model diseles and become the centers of desulfurization. However, naphthalene molecules will resulting the competitive adsorption with DBT molecules if there are naphthalene molecules in the model diseles.

Keywords: CuHY zeolites' absorbent  Dibenzothiophene(DBT)  Desulfurization

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参考文献：

1. CUI Wen-Guang(崔文广), ZHOU Er-Peng(周二鹏), ZHAO Di-Shun(赵地顺), *et al*.. Hebei J. Indu. Sci. & Technol.(河北工业科技)[J], 2004, 21(2): 1—6
2. ZHAO Ye(赵野), SHEN Bao-Jian(申宝剑), GAO Jin-Sen(高金森). Advances in Fine Petrochemicals(精细石油化工进展)[J], 2005, 6(10): 47—53
3. Hernandez-Maldonado A. J., Yang R. T.. J. Am. Chem. Soc.[J], 2004, 126(4): 992—993
4. Mei H., Mei B. W., Yen T. F.. Fuel[J], 2003, 82(4): 405—414
5. ZHAO Di-Shun(赵地顺), REN Hong-Wei(任红威), LI Le(李乐). Chem. J. Chinese Universities(高等学校化学学报)[J], 2007, 28(4): 739—742
6. ZHAO Di-Shun(赵地顺), LIU Cui-Wei(刘翠微), MA Si-Guo(马四国). Chem. J. Chinese Universities(高等学校化学学报)[J], 2006, 27(4): 692—696
7. Abhigeet B. P., Catherine C. L., Brian P. H.. Division of Petroleum Chemistry[J], 2000, 45(4): 689—691
8. Izumi F.. Process for Recovering Organic Sulfur Compounds from Fuel Oil, US 5753102[P], 1998
9. Song C.. Catalysis Today[J], 2003, 86(4): 211—263
10. ZHANG Xiao-Jing(张晓静), QIN Ru-Yi(秦如意), LIU Jin-Long(刘金龙), *et al*.. Petroleum Refinery Engineering(炼油设计)[J], 2001, 31(6): 41— 47
11. Zinnen H. A., Nemeth L. T., Holmgren J. R., *et al*.. Removal of Organic Sulfur Compounds from FCC Gasoline Using Regenerable Adsorbents, US 5843300[P], 1998
12. Yang R. T., Hernandez-Maldonado A. J., Yang F. H.. Science[J], 2003, 301: 79—81
13. Arturo J., Hernandez-Maldonado A. J., Yang R. T., *et al*.. Ind. Eng. Chem. Res.[J], 2004, 43(19): 6142—6149
14. WEI Guo-Xiang(魏国祥), SONG De-Yu(宋德昱), YE Hui-Juan(叶慧娟), et al.. Journal of Fuel Chemistry and Technology(燃料化学学报)[J], 1985, 13(1): 57—64
15. Hidalgo C. V., Itoh H., Hattori T., *et al*.. J. Catal.[J], 1984, 85: 362—369
16. Lobree L. J., Hwang I. Ch., Reimer J. A., *et al*.. J. Catal.[J], 1999, 186: 242—253
17. LI Bin(李斌), LI Shi-Jie(李士杰), LI Neng(李能), *et al*.. Chin. J. Catal.(催化学报)[J], 2005, 26(4): 301—306
18. Roisnel T., Rodriguez-Carvajal J.. Mater. Sci. Forum. European Powder Diffraction Conference (EPDIC 7)[C], 2001, 378—381: 118—123
19. GAO Zi(高滋), HE Ming-Yuan(何鸣元), DAI Yi-Yun(戴逸云), *et al*.. Zeolite Catalysis and Separation Technology(沸石催化与分离技术)[M], Beijing: China Petrochem Press, 1999: 47
20. Yang F. H., Hernandez-Maldonado A. J., Yang R. T.. Sep. Sci. Technol.[J], 39(8): 1717—1732

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1. 谢福中,胡华荣,乔明华,闫世润,范康年,雷浩,谭大力,包信和,宗保宁,张晓昕 .噻吩在猝冷骨架Ni上吸附脱硫的XPS研究[J]. 高等学校化学学报, 2006,27(9): 1729-1732

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