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研究论文

杂原子MCM-41分子筛的合成和催化性能

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摘要:

采用水热合成法合成了金属原子(Zn, Ni, Fe, Al, Cu, Ce)掺杂的MCM-41介孔分子筛(简称T-MCM-41),并将其应用于邻苯二甲酸二(2-乙基己)酯(DOP)的合成反应,研究了不同原子的掺杂对T-MCM-41的结构、比表面积和孔径、酸性及催化性能的影响.结果表明,所制备的T-MCM-41仍然具有六方有序排列结构,比表面积较高(550-900 m²/g)、孔径大(3 nm左右),杂原子的引入使T-MCM-41产生了酸中心,从而使其对DOP的合成具有很好的催化活性和选择性.用T-MCM-41(T=Zn, Fe, Al, Cu)催化DOP的合成反应,在5 h的反应时间内苯酐的转化率可以达到95.5%以上,DOP的选择性可达到96.5%以上.T-MCM-41催化剂具有很好的稳定性,Al-MCM-41在重复使用5次后仍具有较好的催化活性.

关键词: 无机非金属材料 介孔分子筛 MCM-41 邻苯二甲酸二(2-乙基己)酯

Synthesis of MCM-41 mesoporous molecular sieves containing heteroatoms and their catalytic activity

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Abstract:

MCM-41 mesoporous molecular sieves containing heteroatoms (Zn, Ni, Fe, Al, Cu, Ce) (T-MCM-41) were synthesized by direct hydrothermal process and were applied in the synthesis of dioctyl phthalate (DOP) in this paper. The effects of different heteroatoms incorporation on structure, specific surface area and pore volume, acidity and catalytic activity of T-MCM-41 were investigated. The results show that T-MCM-41 which still remains the well-ordered hexagonal mesostructure of MCM-41 has high surface area (550-900 m²/g), large pore diameter (~3 nm). Meanwhile, it has high catalytic activity and selectivity in DOP synthesis because heteroatoms incorporation can produce acid centers. When T-MCM-41 (T=Zn, Fe, Al, Cu) is used as the catalyst, the conversion of phthalic anhydride (PA) reaches above 95.5% and DOP selectivity reaches above 96.5% in 5 h. Moreover, T-MCM-41 has good stability and Al-MCM-41 holds good catalytic activity after being reused five times.

Keywords: inorganic non-metallic materials mesoporous molecular sieve MCM-41 dioctyl phthalate

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参考文献:

- [1] J.S.Beck, J.C.Vartuli, W.J.Roth, M.E.Leonowicz, C.T.Kresge, K.D.Schmitt, C.T-W.Chu,

- D.H.Olson, E.W.Sheppard, S.B.McCullen, J.B.Higgins, J.L.Schlenkert, A new family of mesoporous molecular sieves prepared with liquid crystal templates, *Journal of the American Chemical Society*, 114, 10834(1992) [crossref](#)
- [2] ZHANG Jianguo, JIANG Qingzhe, ZHENG Chengguo, SONG Zhaozheng, KE Ming, Synthesis method and its influence factors of Si-MCM-41, *Journal of China University of Petroleum (Natural Science Edition)*, 32(1), 113(2008)
(张建国, 蒋庆哲, 郑成国, 宋昭峥, 柯明, Si-MCM-41的合成及其影响因素, *中国石油大学学报(自然科学版)*, 32(1), 113(2008))
- [3] MENG Huadong, ZHANG Juntao, DING Liqin, LIANG Shengrong, WANG Xiaoquan, Synthesis and characterization of mesoporous molecular sieve Al-MCM-41, *Journal of Xi'an Shiyou University (Natural Science Edition)*, 22(3), 104(2007)
(孟华东, 张君涛, 丁丽芹, 梁生荣, 王小泉, Al-MCM-41介孔分子筛的合成与表征, *西安石油大学学报(自然科学版)*, 22(3), 104(2007))
- [4] Y.R.Wang, N.C.Lang, A.Tuel, Nature and acidity of aluminum species Al MCM-41 with a high aluminum content (Si/Al=1.25), *Microporous and Mesoporous Materials*, 93, 46 (2006) [crossref](#)
- [5] QIAO Kui, ZHANG Fumin, PAN Duoli, ZHANG Niaofei, JIAN Panming, Synthesis, characterization and catalytic oxidation performance of high Ti-containing Ti-MCM-41 molecular sieves, *Chinese Journal of Inorganic Chemistry*, 24(5), 748(2008)
(乔亏, 张富民, 潘多丽, 张鸟飞, 菅盘铭, 高钛Ti-MCM-41分子筛的合成、表征与催化氧化性能考察, *无机化学学报*, 24(5), 748(2008))
- [6] S.Udayakumar, A.Pandurangan, P.K.Sinha, Para-selective ethylation of phenol with diethyl carbonate over mesoporous Al-MCM-41 molecular sieves, *Applied Catalysis A: General*, 272, 267(2004) [crossref](#)
- [7] M.J.B.Souza, A.S.Araujo, A.M.G. Pedrosa, B.A.Marinkovic, P.M.Jardim, E.M.Jr., Textural features of highly ordered Al-MCM-41 molecular sieve studied by X-ray diffraction, nitrogen adsorption and transmission electron microscopy, *Materials Letters*, 60, 2682 (2006) [crossref](#)
- [8] C.Blanco, C.Pesquera, F.Gonzalez, Synthesis and characterization of MCM-41 with different Si/Al molar ratios and different silicon sources. *Studies in Surface Science and Catalysis*, 154, 432(2004) [crossref](#)
- [9] R.Wojcieszak, S.Monteverdi, M.Mercy, I.Nowak, M.Ziolek, M.M.Bettahar, Nickel containing MCM-41 and Al MCM-41 mesoporous molecular sieves: characteristics and activity in the hydrogenation of benzene, *Applied Catalysis A: General*, 268, 241(2004) [crossref](#)
- [10] V.Parvulescu, B.L.Su, Iron, cobalt or nickel substituted MCM-41 molecular sieves for oxidation of hydrocarbons, *Catalysis Today*, 69, 315(2001) [crossref](#)
- [11] R.R.Sever, R.Alcala, J.A.Dumesic, T.W.Root, Vapor-phase silylation of MCM-41 and Ti-MCM-41, *Microporous and Mesoporous Materials*, 66(1), 53(2003)
- [12] CUI Rong, LIU Xiaoqin, SHAN Jiahui, Preparation of Ce-MCM-41 adsorbent and its performance in model gasoline desulfurization, *Natural Gas Chemical Engineering*, 33(2), 11(2008)
(崔榕, 刘晓勤, 单佳慧, Ce-MCM-41分子筛吸附剂的制备及其在模拟汽油脱硫中的性能, *天然气化工*, 33(2), 11(2008))
- [13] KONG Yan, XU Xinjie, WU Yong, ZHANG Rui, WANG Jun, Effect of promoters on the catalytic activity of MCM-41 with high copper content in benzene hydroxylation, *Chinese Journal of Catalysis*, 29(4), 385(2008)
- [14] C.Mahendiran, P.Sangeetha, P.Vijayan, S.J.Sardhar Basha, K.Shanthi, Vapour phase oxidation of tetralin over Cr and Fe substituted MCM-41 molecular sieves, *Journal of Molecular Catalysis A: Chemical*, 275, 84(2007) [crossref](#)
- [15] S.S.Bhoware, A.P.Singh, Characterization and catalytic activity of cobalt containing MCM-41 prepared by direct hydrothermal, grafting and immobilization methods, *Journal of Molecular Catalysis A: Chemical*, 266, 118(2007) [crossref](#)
- [16] L.F.Chen, J.A.Wang, L.E.Norena, J.Aguilar, J.Navarrete, P.Salas, J.A.Montoya, P.Del Angel, Synthesis and physicochemical properties of Zr-MCM-41 mesoporous molecular sieves and Pt/H3PW12O40/Zr-MCM-41 catalysts, *Journal of Solid State Chemistry*, 180, 2958 (2007) [crossref](#)
- [17] JIANG Pingping, Lu Guanzhong, Development of solid catalyst in catalytic synthesis of DOP, *Chemical Industry and Engineering Process*, 21(5), 328(2002)
(蒋平平, 卢冠忠, 固体酸催化合成邻苯二甲酸二辛酯, *化工进展*, 21(5), 328(2002))
- [18] F.T.Sejidov, Y.Mansoori, N.Goodarzi, Esterification reaction using solid heterogeneous acid catalysts under solvent-less condition, *Journal of Molecular Catalysis A: Chemical*, 240, 186(2005)
- [19] M.Ziolek, I.Nowak, B.Kilos, I.Sobczak, P.Decyk, M.Trejda, J.C.Volta, Template synthesis and characterisation of MCM-41 mesoporous molecular sieves containing various

transition metal elements-TME (Cu, Fe, Nb, V, Mo), Journal of Physics and Chemistry of Solids, 65, 571(2004) [crossref](#)

- [20] S.Ajaikumar, A.Pandurangan, Esterification of alkyl acids with alkanols over MCM-41 molecular sieves: Influence of hydrophobic surface on condensation reaction, Journal of Molecular Catalysis A: Chemical, 266, 1(2007) [crossref](#)
- [21] M.Bhagiyalakshmi, K.Shanmugapriya, M.Palanichamy, B.Arabindoo and V.Murugesan, Esterification of maleic anhydride with methanol over solid acid catalysts: a novel route for the production of heteroesters, Applied Catalysis A: General, 267, 77(2004) [crossref](#)
- [22] DOU Maofeng, J1N Shengming, FULing, CHANG Yan, GUAN Haoyuan, YANG Min, Preparation and structure of mesoporous molecular sieves Ti-MCM-41, Journal of the Chinese Ceramic Society, 36(1), 65(2008)
(豆茂峰, 金胜明, 付英, 常燕, 关豪元, 杨敏, 介孔分子筛Ti-MCM-41的制备和结构, 硅酸盐学报, 36(1), 65(2008))
- [23] R.Luque, J.M.Campelo, D.Luna, J.M.Marinhas, A.A.Romero, NH₄F effect in post-synthesis treatment of Al-MCM-41 mesoporous materials, Microporous and Mesoporous Materials, 84, 11(2005) [crossref](#)
- [24] HUANG Shiyong, WANG Haitao, SONG Yanfen, GAO Wenyi, FAN Lirong, LIAN Piyong, Synthesis of mesoporous MCM-41 molecular sieves containing heteroatoms and study of cyclohexane oxidation, Fine Chemicals, 21(1), 41(2004)
(黄世勇, 王海涛, 宋艳芬, 高文艺, 樊丽荣, 连丕勇, 杂原子MCM-41分子筛的合成及对环己烷氧化的研究, 精细化工, 21(1), 41(2004))
- [25] LI Fuxiang, ZHANG Xiangdi, LI Ruifeng, XIE Kechang, Synthesis and characterization of mesoporous Zr-MCM-41, Journal of Fuel Chemistry and Technology, 32(4), 471(2004)
(李福祥, 张香娣, 李瑞丰, 谢克昌, Zr-MCM-41的合成及其表征, 燃料化学学报, 32(4), 471(2004))
- [26] WU Shujie, HUANG Jiahui, WU Tonghao, SONG Ke, WANG Hongsu, XING Lihong, XU Haiyan, XU Ling, GUAN Jingqi, KAN Qiubin, Synthesis, characterization, and catalytic performance of mesoporous Al-SBA-15 for Tert-butylation of phenol, Chinese Journal of Catalysis, 27(1), 9(2006)

本刊中的类似文章

1. 连肖南 陈鸣才 许凯.使用硅油--水体系制备纳米氢氧化镁[J]. 材料研究学报, 2009,23(6): 663-667
2. 武彩霞 刘罡 方海涛 李峰 史鹏飞.杂质离子对非晶态水合氧化钨电化学超电容性能的影响[J]. 材料研究学报, 2009,23(6): 628-634
3. 康晓雪 田彦文 邵忠宝 袁万颂.掺杂对LiFePO₄电化学性能的影响[J]. 材料研究学报, 2009,23(6): 646-651
4. 代伟 吴国松 孙丽丽 汪爱英.衬底偏压对线性离子束DLC膜微结构和物性的影响[J]. 材料研究学报, 2009,23(6): 598-603
5. 邓福铭 卢学军 刘瑞平 徐国军 陈启武 李文铸.在多壁碳纳米管表面高压生长纳米聚晶金刚石纤维[J]. 材料研究学报, 2009,23(6): 604-609
6. 郑传伟 杨振明 张劲松.反应烧结多孔碳化硅的高温氧化行为[J]. 材料研究学报, 2010,24(1): 103-107
7. 王乙潜 梁文双 G.G.ROSS.二氧化硅基质包埋硅纳米晶的微观结构和发光性能[J]. 材料研究学报, 2009,23(4): 352-356
8. 祝元坤 朱嘉琦 韩杰才 梁军 张元纯.磁控溅射制备SiC薄膜的高温热稳定性[J]. 材料研究学报, 2009,23(4): 410-414
9. 王怀义 刁训刚 王武育 郝维昌 王聪 王天民.基于磁控溅射制备纳米微晶NiOx薄膜的方法[J]. 材料研究学报, 2009,23(4): 426-430
10. 谭兴毅 金克新 赵省贵 陈长乐.真空退火La_{0.7}Sr_{0.3}MnO₃薄膜的光诱导特性[J]. 材料研究学报, 2009,23(4): 395-398