

二氧化硅孔结构对CO氧化用担载型纳米金催化剂的影响

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Impact of silica porosity on the catalytic activity of nanosize gold catalyst for CO oxidation

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摘要 采用三种不同孔结构的二氧化硅材料为载体,应用沉积沉淀法制备担载型纳米金催化剂。以CO催化氧化为模型反应,并结合低温N₂吸附脱附、X射线物相分析、X射线光电子能谱和透射电镜等技术考察三种二氧化硅载体对纳米金催化剂结构和性能的影响。结果表明,催化剂中金纳米颗粒与载体孔结构呈现出良好的对应关系,比表面积大、孔径小且分布均匀的二氧化硅制备的金催化剂颗粒粒径最小,CO氧化活性最高。在18 000 mL/(h·g_{cat})、v(CO)/v(O₂)/v(Ar)=1/21/78的反应条件下,其CO完全转化温度为560 K。

关键词: 金催化剂 一氧化碳氧化 二氧化硅 孔结构

Abstract: Series of nanosize gold catalysts supported by three kinds of silica with different structures were prepared by deposition-precipitation. The CO oxidation was utilized as a probe. The impacts of silica on the structure and catalytic activity were characterized by low-temperature N₂ adsorption/desorption, X-ray diffraction (XRD), X-ray photoelectron spectroscopy (XPS), and transmission electron microscope (TEM). The result turns out that the relationship between the gold nanoparticle size and the support's structure does exist. The higher surface area and the smaller pore size of the silica support corresponding to a smaller gold nanoparticle size and higher activity of CO oxidation. The total CO conversion under 18 000 mL/(h·g_{cat}) with gas ratio of v(CO)/v(O₂)/v(Ar)=1/21/78 is 560 K.

Key words: [gold catalyst](#) [CO oxidation](#) [silica](#) [pore structure](#)

收稿日期: 2012-03-27;

基金资助:

宜宾学院博士科研启动基金(2010B12)。

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引用本文:

徐慧远,罗靖洁,严春蓉等. 二氧化硅孔结构对CO氧化用担载型纳米金催化剂的影响[J]. 燃料化学学报, 2012, 40(11): 1397-1402.

XU Hui-yuan, LUO Jing-jie, YAN Chun-rong et al. Impact of silica porosity on the catalytic activity of nanosize gold catalyst for CO oxidation[J]. J Fuel Chem Technol, 2012, 40(11): 1397-1402.

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- [2] CAO J-L, WANG Y, YU X-L, WANG S-R, WU S-H, YUAN Z-Y. Mesoporous CuO- Fe_2O_3 composite catalysts for low-temperature carbon monoxide oxidation[J]. *Appl Catal B*, 2008, 79(1): 26-34.
- [3] OH S-H, HOFLUND G B. Low-temperature catalytic carbon monoxide oxidation over hydrous and anhydrous palladium oxide powders [J]. *J Catal*, 2007, 245(1): 35-44.
- [4] 王桂英, 廉红蕾, 周文辉, 张文祥, 蒋大振, 吴通好. 氯离子含量对Au/ZnO 催化剂常温CO 氧化性能的影响[J]. *燃料化学学报*, 2001, 29(8): 116-118. (WANG Gui-ying, LIAN Hong-lei, ZHOU Wen-hui, ZHANG Wen-xiang, JIANG Da-zhen, WU Tong-hao. Effect of Cl on the performance of CO oxidation over Au/ZnO catalysts[J]. *Journal of Fuel Chemistry and Technology*, 2001, 29(8): 116-118.)
- [5] HARUTA M, YAMADA N, KOBAYASHI T, IIJIMA S. Gold catalysts prepared by coprecipitation for low-temperature oxidation of hydrogen and of carbon monoxide[J]. *J Catal*, 1989, 115(2): 301-309.
- [6] XU H-Y, LI W-Y, SHANG S-Y, YAN C-R. Influence of MgO contents on silica supported nano-size gold catalyst for carbon monoxide total oxidation [J]. *J Nat Gas Chem*, 2011, 20(5): 498-502.
- [7] 董国利, 王建国, 高荫本, 陈诵英. 二氧化钛负载氧化物催化剂上CO 的氧化反应[J]. *燃料化学学报*, 2000, 28(1): 1-4. (DONG Guo-li, WANG Jian-guo, GAO Yin-ben, CHEN Song-ying. Catalytic activity of CO oxidation of titania-supported oxide catalysts[J]. *Journal of Fuel Chemistry and Technology*, 2000, 28(1): 1-4.)
- [8] SOMODI F, BORBÁTH I, HEGEDUS M, TOMPOS A, SAJÓI E, SZEGEDI Á, ROJAS S, FIERRO J L G, MARGITFALVI J L. Modified preparation method for highly active Au/SiO₂ catalysts used in CO oxidation[J]. *Appl Catal A*, 2008, 347(2): 216-222.
- [9] BORE M T, PHAM H N, SWITZER E E, WARD T L, FUKUOKA A, DATYE A K. The role of pore size and structure on the thermal stability of gold nanoparticles with mesoporous silica[J]. *J Phys Chem B*, 2005, 109(7): 2873-2880.
- [10] OKUMURA M, NAKAMURA S, TSUBOTA S, NAKAMURA T, AZUMA M, HARUTA M. Chemical vapor deposition of gold on Al₂O₃, SiO₂, and TiO₂ for oxidation of CO and of H₂[J]. *Catal Lett*, 1998, 51(1/2): 53-58.
- [11] OVERBURY S H, ORTIZ S L, ZHU H G, LEE B, AMIRIDIS M D, DAI S. Comparison of Au catalysts supported on mesoporous titania and silica: Investigation of Au particle size effects and metal-support interactions[J]. *Catal Lett*, 2004, 95(3/4): 99-106.
- [12] CHOU J, FRANKLIN N R, BAECK S H, JARAMILLO T F, MCFARLAND E W. Gas-phase catalysis by micelle derived Au nanoparticles on oxide supports[J]. *Catal Lett*, 2004, 95(3/4): 107-111.
- [13] BRUNAUER S, EMMETT P H. Chemisorptions of gases on iron synthetic ammonia catalysts[J]. *J Am Chem Soc*, 1940, 62(7): 1732-1746.
- [14] ROUQUEROL J, AUNIR D, FAIRBRIDGE C W, EVEREET D H, HAYNES J H, PERNICONE N, RAMSAY J D F, SING K S W, UNGER K K. Recommendations for the characterization of porous solids[J]. *Pure Appl Chern*, 1994, 66(8): 1739-1758.
- [15] 苏继新, 张慎平, 马丽媛, 屈文, 张明博. Au/SBA-15的制备及其催化 CO 氧化反应性能[J]. *催化学报*, 2010, 31(7): 839-845. (SU Ji-xin, ZHANG Shen-ping, MA Li-yuan, QU Wen, ZHANG Ming-bo. Preparation of Au/SBA-15 and its catalytic activity for CO oxidation [J]. *Chinese Journal of Catalysis*, 2010, 31(7): 839-845.)
- [16] XU H-Y, CHU W-Y, LUO J-J, ZHANG T. Impacts of MgO promoter and preparation procedure on meso-silica supported nano gold catalysts for carbon monoxide total oxidation at low temperature[J]. *Chem Eng J*, 2011, 170(2/3): 419-423.
- [17] ZHANG X, SHI H, XU B-Q. Catalysis by gold: Isolated surface Au³⁺ ions are active sites for selective hydrogenation of 1, 3-butadiene over Au/ZrO₂ catalysts[J]. *Angew Chem Int Ed*, 2005, 44(43): 7132-7135.
- [18] BOCCUZZI F, CHIORINO A, MANZOLI M, LU P, AKITA T, ICHIKAWA S, HARUTA M. Au/TiO₂ nanosized samples: A catalytic, TEM, and FTIR study of the effect of calcination temperature on the CO oxidation [J]. *J Catal*, 2001, 202(2): 256-267.
- [19] XU H-Y, CHU W, LUO J-J, LIU M. New Au/FeOx/SiO₂ catalysts using deposition-precipitation for low-temperature carbon monoxide oxidation[J]. *Catal Commun*, 2010, 11(9): 812-815.
- [20] BOND G C. The effect of the metal to non-metal transition on the activity of gold catalysts[J]. *Faraday Discuss*, 2011, 152: 277-291.
- [21] CHANG C-T, LIAW B-J, HUANG C-T, CHEN Y-Z. Preparation of Au/Mg_xAlO hydroxylate catalysts for CO oxidation[J]. *Appl Catal A*, 2007, 332(2): 216-224.
- [22] LIM D C, LOPEZ-SALIDO I, DIETSCHÉ R, BUBEK M, KIM Y D. Size-selectivity in the oxidation behaviors of Au nanoparticles [J]. *Angew Chem Int Ed*, 2006, 45(15): 2413-2415.
- [23] QIAN K, HUANG W-X, JIANG Z-Q, SUN H-X. Anchoring highly active gold nanoparticles on SiO₂ by CoO_x additive[J]. *J Catal*, 2007, 248 (1): 137-141.
- [1] 谷传涛, 李光俊, 胡蕴青, 庆绍军, 侯晓宁, 高志贤. 淀粉改性SiO₂载体的预处理温度 对其负载的铜基催化剂甲醇转化性能的影响[J]. *燃料化学学报*, 2012, 40(11): 1328-1335.
- [2] 石利红, 李晓峰, 李德宝, 孙予罕. SiO₂ 的甲硅烷基化对钴基催化剂费-托合成催化性能的影响[J]. *燃料化学学报*, 2012, 40(06): 737-742.
- [3] 尹建军, 段钰锋, 王运军, 王卉, 冒咏秋, 韦红旗. 生物质焦的表征及其吸附烟气中汞的研究[J]. *燃料化学学报*, 2012, (04): 390-396.

- [4] 李杨, 廖卫平, 索掌怀. KOH改性对TiO₂结构及其负载金催化剂CO氧化反应活性的影响[J]. 燃料化学学报, 2011, 39(1): 47-53.
- [5] 张谋, 陈汉平, 赵向富, 王贤华, 杨海平, 张世红. 富钙生物油煅烧过程中孔结构变化特性的研究[J]. 燃料化学学报, 2011, 39(06): 443-448.
- [6] 李绍锋, 吴诗勇. 高温下煤焦的碳微晶及孔结构的演变行为[J]. 燃料化学学报, 2010, 38(05): 513-517.
- [7] 于强强, 董园园, 廖卫平, 金明善, 何涛, 索掌怀. CeO₂-Al₂O₃负载金催化剂用于水煤气变换反应的催化活性[J]. 燃料化学学报, 2010, 38(02): 223-229.
- [8] 徐晓玲, 徐秀峰, 张国涛, 牛宪军. 钴铝复合氧化物负载金催化剂的制备及催化分解N₂O[J]. 燃料化学学报, 2009, 37(05): 595-600.
- [9] 黄艳琴, 阴秀丽, 吴创之, 汪从伟, 谢建军, 周肇秋, 马隆龙, 李海滨. 稻秆半焦与CO₂气化反应特性的研究[J]. 燃料化学学报, 2009, 37(03): 289-295.
- [10] 张林仙, 吴晋沪, 王洋. 无烟煤焦气化过程中孔结构的变化及对气化反应性影响的研究[J]. 燃料化学学报, 2008, 36(05): 530-533.
- [11] 黄艳芳, 马正飞, 姚虎卿. 活性炭吸附CO₂与其微孔体积的关系[J]. 燃料化学学报, 2008, 36(03): 343-348.
- [12] 杨帆, 周志杰, 王辅臣, 刘海峰, 龚欣, 于遵宏. 神府煤焦与水蒸气、CO₂气化反应特性研究[J]. 燃料化学学报, 2007, 35(06): 660-666.
- [13] 付志新, 郭占成. 焦化过程半焦孔隙结构时空变化规律的实验研究——孔结构的分形特征及其变化[J]. 燃料化学学报, 2007, 35(04): 385-390.
- [14] 尉迟唯, 李保庆, 李文, 陈皓侃. 煤孔结构特性对水煤浆性质的影响分析[J]. 燃料化学学报, 2006, 34(01): 5-9.
- [15] 胡拖平, 秦张峰, 王建国. SiO₂负载磷钨杂多酸催化的甲苯与乙酸酐酰化反应[J]. 燃料化学学报, 2005, 33(05): 622-625.

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