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研究论文

$LaMO_3$ 纳米复合钙钛矿氧载体化学循环重整甲烷制合成气

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摘要 采用溶胶-凝胶法制备了不同 B 位可变价离子的 LaMO3 (M = Cr, Mn, Fe, Co) 复合氧化物氧载体, 采用 X 射线衍射、N2 吸附-脱附、扫描电镜及 CH4 程序升温表面反应等手段对氧载体进行了表征, 并用于直接选择氧化 CH4 的反应中. 结果表明, Cr, Mn, Fe 和 Co 均能形成 LaMO3 纳米复合钙钛矿结构, 其氧物种氧化能力大小顺序为 LaCoO3 > LaMnO3 > LaFeO3 > LaCrO3. 在连续流动化学循环甲烷重整反应中, LaFeO3 中的氧物种具有更好的选择氧化性能 (H2/CO = 2.06), 其 CH4 转化率和 CO 选择性分别达到 89.6% 和 98.9%. 10 个连续顺序氧化-还原化学循环重整反应中, CH4 转化率约为 60%~70%, CO 选择性达 98% 以上; 且其结构保持了较高的稳定性.

关键词: 甲烷 化学循环重整 钙钛矿 B 位离子 氧载体 合成气

Abstract: Catalytic reforming of natural gas is a commercial process to produce syngas, which is the main source for the production of ammonia, methanol, hydrogen, and many other important products. This method produces also large amounts of CO_2 as by-product. Chemical-looping reforming (CLR) is a novel technology that can be used for syngas production by partial oxidation and steam reforming of hydrocarbon fuels. One key issue with the CLR concept that is being widely studied is the oxidation and reduction behavior of potential oxygen-carrier materials. Four perovskite-based nano-composite oxides were prepared by the sol-gel method and characterized by X-ray diffraction, N_2 adsorption-desorption, scanning electron microscopy (SEM), and CH_4 temperature-programmed surface reaction. The catalytic performance of the prepared samples for CLR of CH_4 to syngas was investigated. The results showed that the $LaMO_3$ (B = Cr, Mn, Fe, and Co) oxides possess perovskite-type nano-composite structure. The oxidizing ability of these four perovskite oxides follows the order of $LaCoO_3$ > $LaMnO_3$ > $LaFeO_3$ > $LaCrO_3$. Among them, $LaFeO_3$ oxide has higher activity for CLR of CH_4 to syngas. The CH_4 conversion and CO selectivity are 89.6% and 98.9%, respectively. Especially, the sequential redox reaction revealed that the $LaFeO_3$ oxide exhibits high stability with CH_4 conversion of 60%~70% and CO selectivity of ~98% after 10 redox cycles. The SEM analysis revealed that the structure of the $LaFeO_3$ oxide was not dramatically changed before and after 10 cyclic reactions.

Keywords: methane, chemical-looping reforming, perovskite, B-site cation, oxygen carrier, syngas

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[1] 小平, 余长春. 化学进展 (Dai X P, Yu Ch Ch. Progr Chem), 2009, 21: 1626

[2] ydén M, Lyngfelt A, Mattisson T. Energy Fuels, 2008, 22: 2585

[3] ydén M, Lyngfelt A, Mattisson T. Fuel, 2006, 85: 1631

[4] ai X P, Yu Ch Ch, Li R J, Wu Q, Hao Z P. J Rare Earth, 2008, 26: 76

[5] ai X P, Li R J, Yu C C, Hao Z P. J Phys Chem B, 2006, 110: 22525

[6] ai X P, Wu Q, Li R J, Yu C C, Hao Z P. J Phys Chem B, 2006, 110: 25856

[7] e Diego L F, Ortiz M, García-Labiano F, Adánez J, Abad A, Gayán P. Energy Procedia, 2009, 1: 3

[8] e F, Wei Y G, Li H B, Wang H. Energy Fuels, 2009, 23: 2095

[9] oghtaderi B, Song H. Energy Fuels, 2010, 24: 5359

[10] Kang K S, Kim C H, Bae K K, Cho W C, Kim S H, Par C S. Int J Hydrogen Energy, 2010, 35: 12246

[11] Rydén M, Lyngfelt A, Mattisson T, Chen D, Holmen A, Björgum E. Int J Greenhouse Gas Control, 2008, 2: 21

[12] Linderholm C, Jerndal E, Mattisson T, Lyngfelt A. Chem Eng Res Des, 2010, 88: 661

[13] de Diego L F, Ortiz M, Adánez J, García-Labiano F, Abad A, Gayán P. Chem Eng J, 2008, 144: 289

[14] Vázquez M I S, Delgado-Vigil M D, Gutiérrez J S, Collins-Martínez V, Ortiz A L. J New Mater Electrochem Systems, 2009, 12: 29

[15] Mihai O, Chen D, Holmen A. Ind Eng Chem Res, 2011, 50: 2613

[16] Evdou A, Zaspalis V, Nalbandian L. Fuel, 2010, 89: 1265

[17] Rida K, Benabbas A, Bouremmad F, Peñ M A, Sastre E, Mart?nez-Arias A. Appl Catal B, 2008, 84: 457

[18] Petrovi? S, Terlecki-Bari?evi? A, Karanovi? L, Kirilov- Stefanov P, Zduji? M, Dondur V, Paneva D, Mitov I, Raki? V. Appl Catal B, 2008, 79: 186

[19] Dacquin J P, Lancelot C, Dujardin C, Da Costa P, Djega-Mariadassou G, Beaunier P, Kaliaguine S, Vaudreuil S, Royer S, Granger P. Appl Catal B, 2009, 91: 596

[20] Wei Z X, Wei L, Gong L, Wang Y, Hu C W. J Hazard Mater, 2010, 177: 554

[21] Ciambelli P, Cimino S, De Rossi S, Lisi L, Minelli G, Porta P, Russo G. Appl Catal B, 2001, 29: 239

[22] Liu B C, Gao L Z, Liang Q, Tang S H, Qu M Z, Yu Z L. Catal Lett, 2001, 71: 225

[1] 黄健, 马人熊, 高志华, 沈朝峰, 黄伟.CeO_2/Ni/Mo/SBA-15 甲烷二氧化碳重整催化剂的表征和催化性能[J]. 催化学报, 2012,33(4): 637-644

[2] 胡全红, 黎先财, 杨爱军, 杨春燕.$BaTiO_3$-$BaAl_2O_4$-Al_2O_3 复合载体的制备、表征及其 Ni 基催化剂催化 CH_4/CO_2 重整反应性能[J]. 催化学报, 2012,33(3): 563-569

[3] 侯玉慧, 常刚, 翁维正, 夏文生, 万惠霖.非水溶剂溶胶-凝胶法制备的纳米卤氧化镧在甲烷氧化偶联反应中的应用[J]. 催化学报, 2011,32(9): 1531-1536

[4] 刘彤, 于琴琴, 王卉, 蒋晓原, 郑小明.等离子体与催化剂协同催化 CH_4 选择性还原脱硝反应[J]. 催化学报, 2011,32(9): 1502-1507

[5] 王月娟, 郭美娜, 鲁继青, 罗孟飞.介孔 Al_2O_3 负载 PdO 催化甲烷燃烧反应性能[J]. 催化学报, 2011,32(9): 1496-1501

[6] 李霞, 杨霞珍, 唐浩东, 刘化章.载体对合成气制甲烷镍基催化剂性能的影响[J]. 催化学报, 2011,32(8): 1400-1404

[7] 张佳瑾, 李建伟, 朱吉钦, 王越, 陈标华.助剂对 Cu-Mn 复合氧化物整体式催化剂催化低浓度甲烷燃烧反应性能的影响[J]. 催化学报, 2011,32(8): 1380-1386

[8] 杨文, 储伟, 江成发, 文婕, 孙文晶.CeO_2 助 Ni/MgO 催化剂用于化学气相沉积法制备多壁碳纳米管[J]. 催化学报, 2011,32(8): 1323-1328

[9] 龙华丽, 胡诗婧, 徐艳, 覃攀, 尚书勇, 印永祥, 戴晓雁.光辐照驱动 CH_4-CO_2 重整中 Ni/MgO-Al_2O_3 催化活性吸收体的活性[J]. 催化学报, 2011,32(8): 1393-1399

[10] 覃攀1, 徐慧远2, 龙华丽1, 冉祎1, 尚书勇2, 储伟1, 印永祥1,*, 戴晓雁1.常压高频放电等离子体炬改进制备 CO_2/CH_4 重整用 Ni/MgO 催化剂[J]. 催化学报, 2011,32(7): 1262-1268

[11] 吕兆坡, 唐浩东, 刘采来, 刘化章*.酸处理活性炭对其负载的 Co-Zr-La 催化剂上 CO 加氢制高碳醇反应性能的影响[J]. 催化学报, 2011,32(7): 1250-1255

[12] 鲁敏 1,2, 吕鹏梅 1,*, 袁振宏 1, 李惠文 1, 许敬亮 1.由杉木锯屑生物质制合成气: 镍基整体式催化剂的表征及催化性能[J]. 催化学报, 2011,32(6): 1017-1021

[13] 马飞, 储伟*, 黄利宏, 余晓鹏, 吴永永.Zn 掺杂的 LaCoO3 钙钛矿用于乙醇水蒸气重整制氢反应[J]. 催化学报, 2011,32(6): 970-977

[14] 高志华, 黄伟, 阴丽华, 谢克昌.La 和 Mn 助剂对完全液相法制备的 CuZr 浆状催化剂结构和性能的影响[J]. 催化学报, 2011,32(2): 309-314

[15] 杨建华, 于素霞, 胡慧晔, 初乃波, 鲁金明, 殷德宏, 王金渠.无第二模板剂法合成多级结构 ZSM-5 分子筛微球及其在甲烷无氧芳构化反应中的应用[J]. 催化学报, 2011,32(2): 362-367