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Article

Mesoporous LaFeO₃ catalysts for the oxidation of toluene and carbon monoxide

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ARTICLE INFO

Article history:

Received 12 June 2013

Accepted 20 August 2013

Published 20 December 2013

Keywords:

Wormhole-like mesoporous LaFeO₃

Perovskite-type oxide

Hard template method

Carbon monoxide oxidation

Toluene oxidation

ABSTRACT

Wormhole-like orthorhombic LaFeO₃ catalysts (LFO-1 and LFO-2) were prepared using the KIT-6 and SiO₂ nanospheres as template, respectively. LFO-1 showed better catalytic activity with $T_{50\%}$ and $T_{90\%}$ of 155 and 180 °C for CO oxidation, and of 200 and 253 °C for toluene oxidation at 20000 mL/(g·h). The excellent catalytic performance was associated with its larger surface area (138 m²/g), higher adsorbed oxygen concentration, and better low-temperature reducibility as well as a wormhole-like mesoporous structure.

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1. Introduction

CO and most volatile organic compounds (VOCs) emitted from industrial and transportation activities are harmful to the atmosphere and human health [1], and it is urgent to control the emissions of CO and VOCs [2]. Catalytic oxidation is one of the most efficient pathways for the removal of CO and VOCs. The critical issue is the availability of an effective catalyst [3,4]. Although supported noble metal catalysts show excellent low temperature activities [5,6], their high costs limit application. Therefore, there is a need for cheap materials with excellent low temperature catalytic performance for CO and toluene oxidation.

Many efforts have been made to study the preparation and catalytic properties of base metal oxides and perovskite-type

oxides (ABO₃). These materials are catalytically active in the oxidation of CO and VOCs. It is well known that a porous catalyst can provide more active sites because of its high surface area. Usually, the preparation of an ABO₃ oxide involves a high temperature solid state reaction, which makes it difficult to preserve a porous structure and large surface area. Therefore, it is highly desirable to establish a strategy for the controlled fabrication of porous ABO₃ oxide with high surface areas. In recent years, the template method has been used to synthesize porous inorganic solids. Among the ABO₃ materials, LaFeO₃ has received much attention in recent years. Wang et al. [7] obtained ordered mesoporous LaCoO₃ using the mesoporous vinyl silica template method. Using a nanocasting strategy with mesoporous silica (KIT-6) as a hard template, Kaliaguine and coworkers [8] prepared wormhole-like mesoporous LaBO₃ (B =

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The work was supported by the National Natural Science Foundation of China (21103005, 21077007), the Natural Science Foundation of Beijing Municipality (KZ200610005004), the Discipline and Postgraduate Education Foundation (PXM2013_014204_07_000261 and 005000542513551), and the Funding Project for Academic Human Resources Development in Institutions of Higher Learning under the Jurisdiction of Beijing Municipality (PHR201107104).

DOI: 10.1016/S1872-2067(12)60689-5 | http://www.sciencedirect.com/science/journal/18722067 | Chin. J. Catal., Vol. 34, No. 12, December 2013

Mn, Co, Fe) with a surface area of 110–155 m²/g and attributed the good catalytic activity of mesoporous LaMnO₃ to its higher surface area.

Our group have prepared three dimensional (3D) ordered mesoporous Co₃O₄ [9], chromia [10,11], and Fe₂O₃ [12] with high surface areas using the KIT-6 and SBA-16 templates, and the results showed that these porous materials had excellent catalytic activities for the oxidation of toluene, methanol, ethyl acetate, formaldehyde, and acetone. In this work, we report the KIT-6 and silica nanosphere template preparation of wormhole-like mesoporous LaFeO₃ catalysts and their catalytic properties for CO and toluene oxidation. Further work on the fabrication and catalytic applications of mesoporous LaCoO₃ and LaMnO₃ is ongoing.

2. Experimental

2.1. Catalyst preparation

The KIT-6 and SiO₂ nanosphere hard templates were synthesized according to procedures described elsewhere [13,14]. The wormhole-like mesoporous LaFeO₃ samples were prepared as follows. Stoichiometric amounts of lanthanum and ferric nitrates and an excess amount of citric acid (lanthanum nitrate/ferric nitrate/citric acid molar ratio = 3.75:3.75:8) were dissolved in 20 mL of methanol and deionized water (methanol/water volumetric ratio = 3:1). After stirring at room temperature (RT) for 8 h to obtain a homogeneous solution, 1.0 g of KIT-6 or SiO₂ nanospheres were added to the above mixed solution at 70 °C under stirring for 4 h. Then, the hard template containing solution was stirred at RT under ultrasonation for 2 h until it was dried. Finally, the dried solid was calcined at a ramp of 2 °C/min in N₂ flow (20 mL/min) from RT to 750 °C and kept at 750 °C for 4 h, then cooled to RT at the same atmosphere, and finally heated at a ramp of 1 °C/min in air flow (20 mL/min) from RT to 750 °C and kept at this temperature for 4 h. To achieve a higher loading, the impregnation, drying, and calcination steps were repeated three times. The silica framework was etched three times at RT using a NaOH aqueous solution (2.0 mol/L). After centrifugation, washing with deionized water and ethanol, and drying at 80 °C overnight, yellow LaFeO₃ powders were obtained. The LaFeO₃ samples prepared using KIT-6 and SiO₂ nanospheres as the hard template were denoted as LFO-1 and LFO-2, respectively. For comparison, we also prepared bulk LaFeO₃ (LFO-bulk) microparticles using the citric acid complexing method after calcination in air at 850 °C for 4 h. All chemicals (A.R.) were purchased from Beijing Chemicals Reagent Company and used without further purification.

2.2. Catalyst characterization

The crystal structure of the catalysts was determined on an X-ray diffractometer (XRD, Bruker/AXS D8 Advance) with Cu K_α radiation and Ni filter ($\lambda = 0.15406$ nm). The operating voltage and current were 40 kV and 35 mA, respectively. The XRD patterns obtained were referred to the JCPDS data for the iden-

tification of the crystal phases. Scanning electron microscope (SEM) images of the samples were taken on a Gemini Zeiss Supra 55 apparatus (operating at 10 kV). Transmission electron microscope (TEM) images as well as the selected area electron diffraction (SAED) patterns of the samples were obtained using a JEOL-2010 equipment (operating at 200 kV). BET surface areas of the samples were measured by N₂ adsorption at –196 °C on a Micromeritics ASAP 2010 analyzer. The samples were degassed under vacuum at 200 °C for 2 h before measurement. The pore size distributions of the samples were calculated using the BJH method.

X-ray photoelectron spectroscopy (XPS, VG CLAM 4 MCD analyzer) was used to determine the La 3d, Fe 2p, O 1s, and C 1s binding energies (BEs) of surface species. Before XPS measurements, the sample was calcined in O₂ (20 mL/min) at 700 °C for 1 h and then cooled to RT, followed by He (20 mL/min) purging at RT for 1 h. After the above treatment, the sample was mounted and transferred to the spectrometer in a transparent glove bag (Instruments for Research and Industry, USA) filled with He to avoid exposure to air. The pretreated sample was degassed in the preparation chamber (10^{–5} Torr) for 0.5 h and then introduced into the analysis chamber (3 × 10^{–9} Torr) for XPS. The C 1s line at BE = 284.6 eV of surface adventitious carbon was taken as a reference for BE calibration.

Hydrogen temperature-programmed reduction (H₂-TPR) experiments were carried out on a chemical adsorption analyzer (Micromeritics Autochem II 2920). Before measurement, 50 mg of catalyst (40–60 mesh) was loaded into a quartz fixed bed U-shaped microreactor (i.d. = 4 mm) and treated in O₂ (30 mL/min) at 500 °C for 1 h. After it was cooled in the same atmosphere to RT and purged by N₂ (50 mL/min) for 0.5 h, the sample was exposed to 5% H₂-95% Ar mixture (50 mL/min) and heated from RT to 800 °C at a ramp of 10 °C/min. The variation in H₂ concentration of the effluent was monitored online by the chemical adsorption analyzer. The reduction peak was calibrated against that of the complete reduction of a standard CuO sample (Aldrich, 99.995%).

2.3. Catalytic evaluation

A continuous flow fixed bed quartz microreactor (i.d. = 4 mm) was employed to evaluate the catalytic activities of the samples at atmospheric pressure for the oxidation of CO and toluene. The reactant feedstocks were: (1) 1 vol% CO + 20 vol% O₂ + He (balance), total flow rate = 100 mL/min, and space velocity (SV) = ca. 20000 mL/(g·h); and (2) 0.1 vol% toluene + 40 vol% O₂ + N₂ (balance), total flow rate = 33.3 mL/min, and SV = ca. 20000 mL/(g·h). In the case of CO oxidation, the inlet and outlet CO concentrations were analyzed online by a gas chromatograph (Shimadzu GC-14C) equipped with a TCD detector and a Carboxen 1000 column for CO analysis. In the case of toluene oxidation, the outlet gases were analyzed online by a gas chromatograph (GC-2010, Shimadzu) equipped with a FID detector and a TCD detector using a Chromosorb 101 column for organic gas separation and a Carboxen 1000 column for permanent gas separation. The balance of carbon in each run was estimated to be 99.5%.

3. Results and discussion

3.1. Crystal structure

Figure 1 shows the XRD patterns of the LFO samples. By comparing to the XRD pattern of a standard LaFeO_3 sample (JCPDS PDF# 74-2203), all the diffraction peaks of each LFO sample in the 2θ range of 10° – 80° were well indexed to the orthorhombic perovskite structure. No impurity phase was detected. In other words, all the LFO samples had a single phase perovskite structure [15,16]. In addition, no XRD signals due to the amorphous SiO_2 was recorded, indicating the complete removal of KIT-6 and silica nanospheres. There was a difference in the peak intensities of the LFO samples, suggesting differences in crystallinity, which was probably due to the different preparation methods.

3.2. Morphology, pore structure, and surface area

Figure 2 shows the TEM images and SAED patterns of the LFO samples. These samples displayed a wormhole-like mesoporous structure, in agreement with other researchers [8]. In Fig. 2(b) and (d), one can see well resolved lattice fringes. The lattice spacings (d values) of the (112) plane of LFO-1 and LFO-2 were estimated to be 0.275–0.277 nm, close to that

Fig. 1. XRD patterns of LFO-1 (1), LFO-2 (2), and LFO-bulk (3) samples.

Fig. 2. TEM images and SAED patterns (insets) of LFO-1 (a, b) and LFO-2 (c, d) samples.

(0.2780 nm) of the standard LaFeO_3 sample. The recording of multiple bright electron diffraction rings in the SAED patterns indicated that LFO-1 and LFO-2 were polycrystalline. It is worth pointing out that the sample from the KIT-6 hard template exhibited a 3D ordered mesoporous architecture with a surface area of $789 \text{ m}^2/\text{g}$ and an average pore size of 6.2 nm, while that from the SiO_2 nanosphere hard template possessed a surface area of $192 \text{ m}^2/\text{g}$.

Figure 3 shows the N_2 adsorption isotherms and pore size distributions of the LFO samples. Their textural parameters are summarized in Table 1. The LFO-1 sample exhibited a type IV isotherm with a type H1 hysteresis loop in the relative pressure (p/p_0) range of 0.4–0.9 [7,17], which is characteristic of mesoporous materials. The LFO-2 sample displayed a distorted type IV isotherm with a type H3 hysteresis loop at $p/p_0 = 0.4$ –0.9 (Fig. 3(a)), which indicated that there were irregular mesopores in LFO-2. The pore size distributions of the LFO samples

Fig. 3. N_2 adsorption isotherms (a) and pore size distributions (b) of LFO-1 (1) and LFO-2 (2) samples.

Table 1

Textural parameters of the LFO samples.

Sample	Hard template	BET surface area (m ² /g)	Average pore size (nm)	Pore volume (cm ³ /g)
LFO-1	KIT-6	138	4.2	0.196
LFO-2	SiO ₂ nanosphere	65	5.6	0.116
LFO-bulk	—	3	—	—

confirmed these deductions. For each LFO, there was one peak at the pore size range of 3 to 4 nm (Fig. 3(b)). The LFO-1 and LFO-2 samples possessed surface areas of 134 and 65 m²/g, respectively, with pore sizes and pore volumes of 4.2 and 5.6 nm and 0.116 and 0.196 cm³/g, respectively.

3.3. Surface composition, iron oxidation state, and surface oxygen species

Figure 4 illustrates the Fe 2p_{3/2} and O 1s XPS spectra of the LFO samples. Their surface compositions are summarized in Table 2. It can be seen that the asymmetrical Fe 2p_{3/2} signal can be decomposed into two components at BE = 710.1 and 712.1 eV, assignable to surface Fe³⁺ and Fe⁴⁺ species [18]. The surface Fe⁴⁺/Fe³⁺ molar ratios (0.828–0.904) of the porous LFO samples were much higher than that (0.650) of LFO-bulk (Table 2), with the LFO-1 sample showing the highest (0.904). In Fig. 4(b), one can observe an asymmetrical O 1s signal that can be decomposed into three components at BE = 529.5, 531.1, and 533.1 eV, attributable to surface lattice oxygen (O_{latt}), adsorbed oxygen (O_{ads}), and surface adsorbed water species [19], respectively. The surface O_{ads}/O_{latt} molar ratios of LFO-1 (3.42) and

LFO-2 (3.15) were significantly higher than that (1.07) of LFO-bulk. The XPS results revealed that the LFO-1 sample exhibited the highest Fe⁴⁺ and O_{ads} concentrations on the surface.

3.4. Reducibility

Figure 5(a) illustrates the H₂-TPR profiles of the LFO samples. For each sample, the reduction process can be divided into low temperature (≤ 500 °C) and high temperature (> 500 °C) reduction regions. The former was ascribed to the reduction of Fe⁴⁺ to Fe³⁺ as well as the removal of oxygen adspecies, and the latter to the reduction of Fe³⁺ to Fe²⁺ [20,21]. It was noted that the Fe species might not be reduced to metallic Fe⁰ in the range of 500–800 °C [22,23]. The onset and maximum reduction temperatures were different from sample to sample. The maximum temperature of the first reduction peak of LFO-1 or LFO-2 was lower than that of LFO-bulk, suggesting that the porous LFO samples were more reducible than the bulk counterpart. By quantitatively analysis of the H₂-TPR profiles (Table 2), the H₂ consumption amounts of LFO-bulk, LFO-1, and LFO-2 in the RT–500 °C range were 0.29, 2.31, and 1.24 mmol/g_{cat}, and that in the 500–800 °C region were 0.61, 2.07, and 2.73 mmol/g_{cat}, respectively. The LFO-1 sample showed the highest low-temperature H₂ consumption. The low-temperature reducibility can be conveniently evaluated using the initial (where less than 25% oxygen for the first reduction peak of the sample was consumed) H₂ consumption rate of a catalyst [24], as shown in Fig. 5(b). The initial H₂ consumption rate increased in the order of LFO-bulk < LFO-2 < LFO-1. This trend was in good agreement with the sequence in catalytic performance of the samples (shown below).

Fig. 4. Fe 2p_{3/2} (a) and O 1s (b) XPS spectra of the LFO-1 (1), LFO-2 (2), and LFO-bulk (3) samples.

Table 2 Surface composition, H₂ consumption, and CO and toluene oxidation activities of the LFO catalysts.

Catalyst	Surface element composition		H ₂ consumption (mmol/g _{cat})		CO oxidation activity ^a			Toluene oxidation activity ^b		
	Fe ⁴⁺ /Fe ³⁺ molar ratio	O _{ads} /O _{latt} molar ratio	≤ 500 °C	> 500 °C	T _{10%} (°C)	T _{50%} (°C)	T _{90%} (°C)	T _{10%} (°C)	T _{50%} (°C)	T _{90%} (°C)
LFO-bulk	0.650	1.07	0.29	0.61	210	240	310	300	360	420
LFO-1	0.904	3.42	2.31	2.01	125	155	180	130	200	253
LFO-2	0.828	3.15	1.24	2.73	135	170	208	152	210	290

^a Reaction conditions: CO 1 vol%, O₂ 20 vol%, SV = 20000 mL/(g·h);

^b Reaction conditions: toluene 0.1 vol%, O₂ 40 vol%, SV = 20000 mL/(g·h).

Fig. 5. H₂-TPR profiles (a) and initial H₂ consumption rate as a function of inverse temperature (b) of LFO-bulk (1), LFO-1 (2), and LFO-2 (3) samples.

Fig. 6. CO (a) and toluene (b) conversion as a function of temperature over the LFO catalysts at CO concentration = 1 vol% or toluene concentration = 0.1 vol% and SV = 20000 mL/(g·h), and effect of SV on the catalytic activity of LFO-1 for CO (c) and toluene (d) oxidation.

3.5. Catalytic performance

In the blank quartz sand experiment, no significant conversion of CO below 300 °C or toluene below 450 °C was detected, indicating that homogeneous reactions were negligible under the adopted conditions. Figure 6(a) and (b) show the catalytic performance of the LFO samples for CO and toluene oxidation, respectively. CO and toluene conversion increased with temperature, and the catalytic activity was in the order of LFO-bulk < LFO-2 < LFO-1, which was in agreement with the sequences in O_{ads} species concentration and low temperature reducibility.

It was noted that in our catalytic system no incomplete oxidation product was detected. It is convenient to use $T_{10\%}$, $T_{50\%}$, and $T_{90\%}$ (the temperatures required for achieving CO or toluene conversion = 10%, 50%, and 90%, respectively) for activity comparison. As listed in Table 2, the wormhole-like mesoporous LFO-1 sample exhibited the best catalytic activity: the $T_{10\%}$, $T_{50\%}$, and $T_{90\%}$ values were 125, 155, and 180 °C for CO oxidation, and 130, 200, and 253 °C for toluene oxidation.

Figure 6(c) and (d) shows the effect of SV on the catalytic activity of the LFO-1 sample for CO and toluene oxidation, respectively. As expected, the catalytic activity decreased with SV. We also monitored CO and toluene conversion over LFO-1 for

Fig. 7. CO or toluene conversion as a function of on-stream reaction time over the LFO-1 catalyst under the conditions of CO concentration = 1 vol%, O₂ concentration = 20 vol%, SV = 20000 mL/(g·h), and 174 °C or toluene concentration = 0.1 vol%, O₂ concentration = 40 vol%, SV = 20000 mL/(g·h), and 253 °C.

100 h reaction at 174 °C for CO oxidation and at 253 °C for toluene oxidation. The results are shown in Fig. 7. No significant loss in catalytic activity was detected during the reactions. In other words, the LFO-1 sample was catalytically stable under the adopted conditions.

For CO oxidation at SV = 20000 mL/(g·h), the LFO-1 catalyst ($T_{50\%}$ = 155 °C and $T_{90\%}$ = 180 °C) performed much better than porous LaFeO₃ ($T_{50\%}$ = 240 °C and $T_{90\%}$ = 300 °C) [25]. For toluene oxidation at SV = 20,000 mL/(g·h), the catalytic activity ($T_{50\%}$ = 200 °C and $T_{90\%}$ = 253 °C) of LFO-1 was much better than that of porous SrFeO_{3-δ} ($T_{50\%}$ = 292 °C and $T_{90\%}$ = 340 °C) [26], and LaFeO₃ ($T_{50\%}$ = 297 °C and $T_{90\%}$ = 308 °C) [27]; it was similar to that of porous LaMnO₃ ($T_{50\%}$ = 222 °C and $T_{90\%}$ = 243 °C) [28], but it was inferior to that of Pd/LaFeO₃ ($T_{50\%}$ = 139 °C and $T_{90\%}$ = 230 °C) [21].

The catalytic performance of an ABO₃ oxide is related to its surface area, adsorbed oxygen concentration, and reducibility. A higher surface area is beneficial for the catalytic activity [29,30]. The formation of a porous structure facilitates this. More adsorbed oxygen species gives higher catalytic activity [31–33]. The oxidation of CO or toluene over LFO proceeds according to the suprafacial mechanism: CO or toluene first reacts with oxygen adspecies to give CO₂ and water, and meanwhile O₂ molecules are activated to be the active oxygen adspecies by a Fe⁴⁺ ⇌ Fe³⁺ redox process. The strong redox capability of LFO guarantees the recyclability of Fe ions that have two oxidation states, thus facilitating the oxidation of CO or toluene. Compared to LFO-bulk, porous LFO-1, and LFO-2 have much higher surface area and oxygen adspecies concentration and much better low temperature reducibility. These unique characteristics of porous LFO gave them better performance in catalyzing the oxidation of CO or toluene. Therefore, it is concluded that the excellent catalytic performance of LFO-1 was associated with its high surface area and oxygen adspecies concentration, good low temperature reducibility, and wormhole-like mesoporous structure.

4. Conclusions

Porous wormhole-like orthorhombic LaFeO₃ perovskites LFO-1 and LFO-2 were prepared using the KIT-6 and SiO₂ nanosphere templates and possessed surface areas of 138 and 65 m²/g, respectively. The LFO-1 sample showed the better catalytic performance: the $T_{10\%}$, $T_{50\%}$, and $T_{90\%}$ were 125, 155, and 180 °C for CO oxidation, and 130, 200, and 253 °C for toluene oxidation at SV = 20000 mL/(g·h). The large surface area, high oxygen adspecies concentration, good low temperature reducibility, and wormhole-like mesoporous structure were responsible for the excellent catalytic performance of LFO-1.

Acknowledgement

We thank Prof. Chak Tong Au (Hong Kong Baptist University) for doing the XPS analysis.

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Graphical Abstract

Chin. J. Catal., 2013, 34: 2223–2229 doi: 10.1016/S1872-2067(12)60689-5

Mesoporous LaFeO₃ catalysts for the oxidation of toluene and carbon monoxide

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The wormhole-like mesoporous LaFeO₃ samples with surface areas of 65–138 m²/g were prepared by the hard-template method and showed high catalytic activity for the complete oxidation of CO and toluene, which is due to the large surface area, high adsorbed oxygen concentration, and good low-temperature reducibility.

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甲苯和一氧化碳氧化用介孔LaFeO₃催化剂

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摘要: 以KIT-6和二氧化硅纳米球为硬模板制备了比表面积分别为138和65 m²/g的正交相蠕虫状介孔LaFeO₃催化剂LFO-1和LFO-2. 在空速为20000 mL/(g·h)的条件下, LFO-1样品显示出最高的催化活性: 对于CO氧化反应, *T*_{50%}和*T*_{90%}分别为155和180 °C; 对于甲苯氧化反应, *T*_{50%}和*T*_{90%}分别为200和253 °C. 该样品优异的催化性能与它具有较大的比表面积、较高的氧吸附物种浓度、较好的低温还原性以及良好的蠕虫状介孔结构有关.

关键词: 蠕虫状介孔LaFeO₃; 钙钛矿型氧化物; 硬模板合成法; 一氧化碳氧化; 甲苯氧化

收稿日期: 2013-06-12. 接受日期: 2013-08-20. 出版日期: 2013-12-20.

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基金来源: 国家自然科学基金(21103005, 21077007); 北京市自然科学基金B类重点(KZ200610005004); 学科与研究生教育基金(PXM2013_014204_07_000261, 005000542513551); 北京市教委创新团队项目(PHR201107104).

本文的英文电子版由Elsevier出版社在ScienceDirect上出版(<http://www.sciencedirect.com/science/journal/18722067>).