Catalysis Today 265 (2016) 210–217

Contents lists available at ScienceDirect

Catalysis Today

journal homepage: www.elsevier.com/locate/cattod

One-pot synthesis of perovskite-type metal oxides via confined mesopore and their catalytic activity for toluene oxidation

Myung-Ji Suh[a], Young-Kwon Park[b], Son-Ki Ihm[a,*]

[a] Department of Chemical and Biomolecular Engineering, Korea Advanced Institute of Science and Technology, Daejeon 305-338, Republic of Korea
[b] School of Environmental Engineering, University of Seoul, Seoul 130-743, Republic of Korea

ARTICLE INFO

Article history:
Received 30 June 2015
Received in revised form 25 August 2015
Accepted 26 August 2015
Available online 3 November 2015

Keywords:
Perovskite
Mesoporous structure
Toluene oxidation
Surface area

ABSTRACT

One-pot synthesis via confined mesopore using a citrate-based metal precursor was applied to obtain a perovskite-type metal oxide with a high surface area for total oxidation of toluene. The method produced $LaCoO_3$ with a higher surface area (147 m^2/g) and smaller crystallite size (8 nm) than conventional methods, such as the co-precipitation and citrate sol–gel method. The citrate-based metal precursors were inserted in the framework of mesopore and the crystallite size of $LaCoO_3$ converted from these precursors could be adjusted with the confinement effect by mesoporous structure formed with $LaCoO_3$ simultaneously. The catalytic activity for the total oxidation of toluene over $LaCoO_3$ showed a good correlation with the high surface area, which was confirmed by characterization of the chemical states of $LaCoO_3$.

1. Introduction

Reduction of volatile organic compounds (VOCs) included in air pollutants is a major concern on account of increasingly environmental regulations and more stringent air quality standards. The catalytic oxidation of VOCs has been identified as one of the most efficient ways of removing VOCs, because the catalysts allow a considerable decrease in the oxidation temperature compared to thermal oxidation [1–5]. Noble metal-based catalysts are used widely in the catalytic oxidation process owing to their high activity at low temperature [2]. However, noble metal-based catalysts are expensive and less thermally stable.

Lanthanum based perovskite-type metal oxide (ABO_3) catalysts have been suggested as an alternative to noble metals in hydrocarbon oxidation and exhaust removal [6–12,14–22] because of their higher thermal stability and lower cost. On the other hand, the use of perovskite-type metal oxide as a catalyst is limited because of its low surface area. Perovskite-type metal oxides are normally prepared using conventional methods, such as co-precipitation and sol–gel method. In the conventional methods, a high calcination temperature (>800 °C) is necessary to form a perovskite structure and hence the perovskite-type metal oxide has a low surface area (<10 m^2/g) due to sintering [23].

Many studies have reported that an increase in the surface area of perovskite-type metal oxide through a modification of the synthesis method could enhance the catalytic activity for total oxidation [6–22]; Weng et al. [6] synthesized $LaCoO_3/MgO$ with a high surface area of 35.3 m^2/g using a continuous hydrothermal flow synthesis route with supercritical water and reported that it was more effective for toluene oxidation than the catalyst prepared via the citrate route. Pang et al. [7] showed that the catalytic performance of CO oxidation could also be improved by increasing the surface area (from 6.3 m^2/g to 64.1 m^2/g) of $LaCoO_3$ catalysts prepared using the modified sol–gel method. Mesoporous materials have been used widely as good support or template for perovskite-type catalysts because of the huge surface area of the pore structure [20–22].

One-pot synthesis was reported in our previous work [24] showing that copper oxide and mesoporous silica could be formed simultaneously in a single batch reactor and hence various resource was economized more simply than by the conventional impregnation method. The optimal synthetic conditions for the one-pot synthesis were investigated. The way of copper catalyst preparation using the one-pot synthesis method based on mesoporous structure, SBA-15 and MCM-41, respectively, was named as SBA route and MCM route. Especially the SBA route using Cu citrate sol precursor in acidic environment produced a small crystallite size and high surface area of copper oxide.

In this present study, it was attempted to prepare a perovskite-type catalyst with improved catalytic performance for VOCs removal. $LaCoO_3$, as a perovskite-type metal oxide, was synthesized

---

* Corresponding author.
E-mail address: skihm@kaist.ac.kr (S.-K. Ihm).

http://dx.doi.org/10.1016/j.cattod.2015.08.057

Table 1
Physical properties of prepared catalysts.

Sample	Crystal phase	Surface area[a] (m^2/g)	Pore volume[a] (cm^3/g)	Avg. pore diameter[b] (nm)	Porewall thickness (nm)	Crystallite size[c] (nm)
SBA-15	SBA-15	1050	1.1	5.4	3.0	
LCO-m	$LaCoO_3$ and SBA-15	841	0.6	3.3	3.4	
LCO	$LaCoO_3$	147	0.1			8
LCO-cp	$LaCoO_3$	6	0.01			69
LCO-ct	$LaCoO_3$	10	0.01			55

[a] Estimated by using BET method from nitrogen adsorption isotherm.
[b] Estimated by using BJH method from nitrogen desorption isotherm.
[c] Estimated by Scherrer equation.

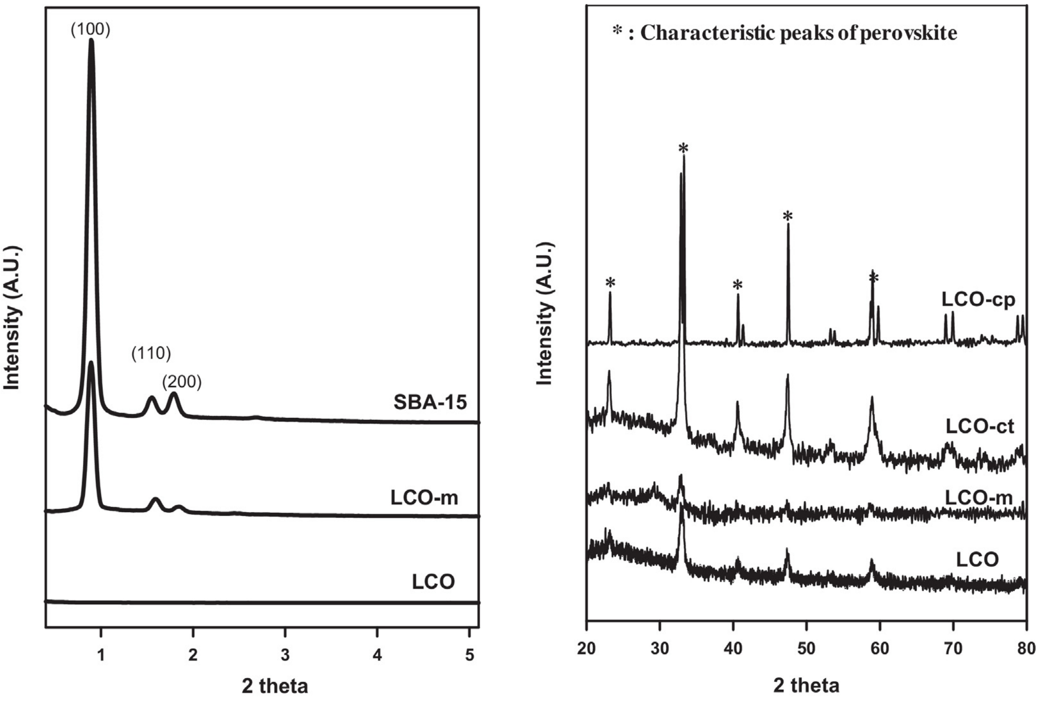

Fig. 1. XRD patterns of catalysts prepared by one-pot synthesis (LCO-m and LCO), catalysts prepared by conventional methods (LCO-ct and LCO-cp), and mesoporous silica (SBA-15).

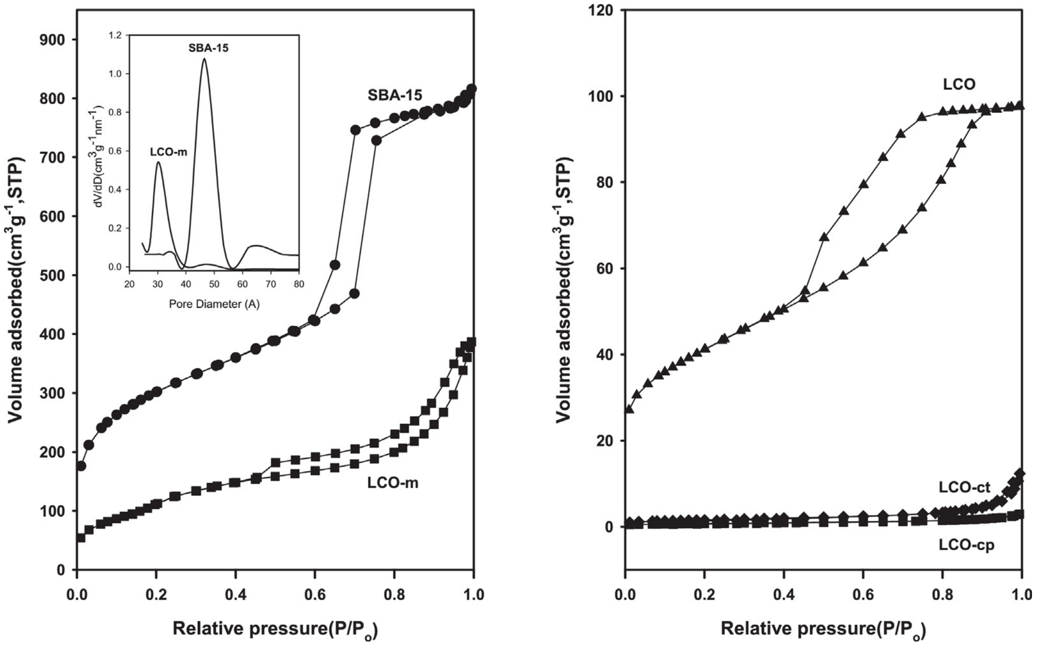

Fig. 2. N_2-adsorption/desorption isotherms of catalysts prepared by one-pot synthesis (LCO-m and LCO), catalysts prepared by conventional methods (LCO-ct and LCO-cp), and mesoporous silica (SBA-15).