



Synthesis of nano-crystalline $\text{La}_{1-x}\text{Sr}_x\text{CoO}_{3-\delta}$ perovskite oxides by EDTA–citrate complexing process and its catalytic activity for soot oxidation

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ABSTRACT

In the present study, $\text{La}_{1-x}\text{Sr}_x\text{CoO}_{3-\delta}$ ($x = 0.2, 0.3, 0.4, 0.5$) perovskite nano-particles have been successfully prepared by EDTA–citrate complexing process and the catalytic performance was examined for the soot oxidation activity. The physio-chemical properties of these materials were characterized by means of X-ray diffraction (XRD), thermogravimetry–differential thermal (TG–DTA) analysis, BET surface area and X-ray photoelectron spectroscopy (XPS) analysis, scanning electron microscopy/energy dispersive spectroscopy (SEM/EDS) analysis. TG–DTA analysis suggested that, in the temperature range of 350–500 °C, the pyrolysis of metal/nitrate/citrate/EDTA complexes takes place and the single phase $\text{La}_{1-x}\text{Sr}_x\text{CoO}_{3-\delta}$ perovskite oxides can be formed at 900 °C and the phase formation has been confirmed by XRD analysis. The surface composition of the samples derived from XPS analysis indicated an impoverishment of lanthanum content at the surface with the increase of strontium content. For $\text{La}_{0.6}\text{Sr}_{0.4}\text{CoO}_{3-\delta}$ sample, Sr-enriched perovskite surface has been observed along with the more number of surface oxides (M(Sr, La, Co)–O) resulting in higher catalytic activity for soot oxidation than the others.

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1. Introduction

Solid oxide fuel cells (SOFCs) are the promising energy production systems because of their high efficiency, environmental friendliness and utilization of a variety of the fuel resources [1–7]. In the recent years, perovskite type oxides have received much attention due to their potential application as SOFC electrode materials, solid oxide electrolysis cell (SOEC) contact layers between oxygen electrode and interconnects, gas sensors, oxygen permeable membranes and catalyst for steam and hydrocarbon reforming [8–12]. Some of the advantages with these oxides are (i) the basic structure can be retained unaltered with variation in composition and constituent elements and (ii) valence state of the individual constituents, stoichiometry of the oxide and the defects/vacancies can be varied widely which in turn tailor the physical, chemical and transport properties of these oxides [13,14]. Generally, the functional properties of the perovskite oxides are influenced by synthesis methods, calcination conditions (temperature, time and atmosphere) and A and/or B site substitution in the ABO_3 perovskite structure [13–16]. Partial substitution of A-site cations by proper cations e.g. lanthanum by strontium [17,18] can alter the transport properties of perovskite oxides. Substitutions in A and/or B sites

create defects and hence modify the catalytic and other functional properties [19,20].

Strontium-substitutes lanthanum cobalt oxides ($\text{La}_{1-x}\text{Sr}_x\text{CoO}_{3-\delta}$) with the perovskite structure have been great scientific interest because of their magnetic behavior associated with a spin state [21,22], high catalytic activity for oxidation [23,24] and excellent performance as an oxygen electrode [25,26]. One of the main challenges in developing perovskite catalysts is to obtain their structures while preserving sufficiently high surface areas. Generally, the preparation of a perovskite oxide involves a solid-state reaction of its precursor oxide to form characteristic ABO_3 structure. This requires a significant exposure of the precursor oxide to high temperatures, thus leading to a low specific surface area of the catalyst. These perovskite oxides were prepared by various synthesis methods like Pechini gel technique [27], solid state reaction [28], glycine nitrate process [29], microwave process [30] and the EDTA methods [31] in order to overcome this limitation have been tested.

Among the various synthesis methods, EDTA–citrate complexing process is a modified Pechini type reaction, is a polymerized complex method to synthesize a wide variety of oxide materials [17,32]. Various metal ions in a solution are chelated to form metal complexes and then these chelates undergo poly-esterification when heated to form a polymeric glass which has metal ions uniformly distributed throughout. Thereby, the resultant powders have the nano-scaled particle size and nice compositional homogeneity at molecular level.

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