



Advanced $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Zn}_{0.2}\text{Fe}_{0.8}\text{O}_{3-\delta}$ perovskite-type ceramics as oxygen selective membranes: Evaluation of the synthetic process

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Abstract

A synthetic route for advanced perovskites is elucidated that consists of a combined EDTA/citrate complexing of metal cations and the subsequent drying and firing at moderate temperatures. A fine-scale intermixing of cations is maintained during all the processing steps. The perovskite-type oxide is formed already in an intermediate step at 700 °C by the reaction of an ultra-finely dispersed powder consisting of a mixed barium–strontium carbonate, a zinc–iron spinel, and zinc oxide. The process yields powders of good sinterability and finally dense ceramics composed of stoichiometric perovskite grains of the type $(\text{Ba,Sr})(\text{Zn,Fe})\text{O}_{3-\delta}$.

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1. Introduction

The economic separation of oxygen from air by membrane technologies stimulates nowadays an emerging field of research and engineering which has an impact on a variety of applications. These span from the decentralized production of oxygen enriched air for medical purposes or for industrial combustion processes over fuel cells for power generation to the production of synthesis gas [1,2]. Our group is engaged in the development of membrane reactors in disk or tubular geometries for the production of synthesis gas by steam reforming [3] or the partial oxidation of methane [4], as well as in the production of oxygen enriched air [5].

Oxygen is transferred from a given chemical potential (high μ_{O_2} , feed side) over an oxygen semi-permeable membrane to a lower chemical potential (low μ_{O_2} , permeate side). Compared with polymeric membranes, membranes made from solid state electrolytes based on oxygen conducting oxides are 100% selective for oxygen. Besides oxygen, the polymeric membranes transport remarkable amounts of noble or inert gases like Ar or CO_2 which can be very disadvantageous for some of the mentioned applications. Under the oxygen conducting ceramics yttria-stabilized ZrO_2 (YSZ) is a widely used material (e.g. oxygen lambda sensor) [6]. But its low electron conductivity and the high operation temperature (1000 °C) make it by far not a suitable membrane material for oxygen separation purposes.

Much more attractive materials are perovskite-type oxides of stoichiometry ABO_3 that can host different cations on the A- or B-site. Their properties can be tuned over a wide range, so conductivities from almost pure electronic to almost pure ionic can be realized. To establish high oxygen permeation fluxes it is essential to set up the ambipolar diffusion of electronic charge carriers. That means both are needed, high ionic and electronic conductivities. A specific acceptor or donor doping on both lattice sites yields oxides of the type $(\text{A,A}')(\text{B,B}')\text{O}_{3-\delta}$. By substitution of acceptors onto either the A- or B-sites high concentrations of oxygen vacancies can be induced (marked by δ in the chemical formula), and thus improved ionic conductivities are obtained. Moreover, lower operational temperatures (800–900 °C) than for YSZ are obtainable [7].

In the pioneering work of Teraoka and co-workers highly A- and B-site doped $(\text{La,Sr})(\text{Co,Fe})\text{O}_{3-\delta}$ perovskites were obtained by the citrate process [8–10]. As these oxides showed conductivities that were two orders of magnitude higher than those observed for YSZ, many researchers were stimulated to investigate related stoichiometries. Despite their excellent electronic and ionic conductivities, all these oxides showed poor stability under low oxygen partial pressures ($P_{\text{O}_2} < 10^{-5}$ Pa). This is mainly due to cobalt leaving the structure easily at high temperatures, especially in reducing atmospheres.

The development of suitable membrane materials with both high ionic (for oxygen) and electronic conductivities that show a good stability in low oxygen potentials, μ_{O_2} , as well as

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