^{57}Fe Mössbauer *in situ* study into oxygen vacancy disorder of BSCF, $(Ba_{0.5}Sr_{0.5})(Co_{0.8}Fe_{0.2})O_{3-\delta}$

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Abstract

Although air is usually used for fuel combustion, it is well known that oxygen enrichment of combustion air enhances the combustion efficiency. Cryogenic oxygen separation is well established for oxygen production at large scale but its costs are relatively high compared to the economic benefit caused by an improved combustion efficiency. Therefore, the development of alternative oxygen separation processes is still an issue. One of the most promising ceramic materials for oxygen separation membranes is the mixed ionic electronic conducting $(Ba_{0.5}Sr_{0.5})(Co_{0.8}Fe_{0.2})O_{3-\delta}$ (BSCF) with its extremely high oxygen vacancy concentration. For 900 °C, for example, a value of $\delta = 0.8$ has been reported at an oxygen partial pressure of 10^{-3} bar [1,2].

We report on ^{57}Fe Mössbauer *in situ* studies of the mixed ionic electronic conducting (MIEC) oxide functional materials $(Ba_{0.5}Sr_{0.5})(Co_{0.8}Fe_{0.2})O_{3-\delta}$, BSCF, conducted between room temperature and 1000 °C in atmospheres of variable oxygen content in order to obtain insight into local coordination and valence of iron at working conditions and into the distribution of oxygen vacancies on their different sites.

The magnetically-split room-temperature Mössbauer spectra of BSCF reveal the presence of two inequivalent iron species [3]. Evaluation of signal intensities confirms results from theoretical computations on vacancy formation in BSCF which indicate that formation energies of the various types of oxygen vacancies differ by the order of 0.1 eV only [4,5]. The analysis also shows that the distribution of vacancies is far from random [3]. In the paramagnetic high-temperature phase ($T \geq$ 315 °C), the quadrupole-split signals demonstrate that local symmetry at iron sites is lower than cubic. At 700, 850, and 1000 °C, Mössbauer centre shifts as well as quadrupole splittings are discussed in respect to stoichiometry-related changes in valence and local coordination of the iron probes.

Recent Publications

1. Yaremchenko AA, Khalyavin DD, Patrakeev MV (2017) Uncertainty of oxygen content in highly nonstoichiometric oxides from neutron diffraction data: example of perowskite-type $(Ba_{0.5}Sr_{0.5})(Co_{0.8}Fe_{0.2})O_{3-\delta}$. J. Mater. Chem. A. 5:3456-3463

2. Tomkiewicz AC, Tamimi MA, Huq A, McIntosh S (2013) Evidence for the low oxygen stoichiometry of cubic $Ba_{0.5}Sr_{0.5}Co_{0.5}Fe_{0.5}O_{3-\delta}$ from in-situ neutron diffraction. Solid State Ionics 253:27-31

3. Gaczynski P, Harpf A, Boer J, Kircheisen R, Kriegel R, Becker K-D (2018) ^{57}Fe Mössbauer study into oxygen vacancy disorder of $(Ba_{0.5}Sr_{0.5})(Co_{0.8}Fe_{0.2})O_{3-\delta}$. Solid State Ionics 316C:59-65

4. Kotomin EA, Mastrikov YA, Kuklja MM, Merkle R, Roytburd A, Maier J (2011) First principles calculation of oxygen vacancy formation and migration in mixed conducting $(Ba_{0.5}Sr_{0.5})(Co_{1-y}Fe_y)O_{3-\delta}$ perovskites. Solid State Ionics 188:1-5

5.Mastrikov YA, Kuklja MM, Kotomin EA, Maier J (2010) First-principles modelling of complex perovskite $(Ba_{0.5}Sr_{0.5})(Co_{1-y}Fe_y)O_{3-\delta}$ for oxide fuel cell and gas separation membrane applications. Energy Environ. Sci. 3:1544-1550



Biography

Klaus D. Becker studied Physics at the University of Göttingen, Germany, where he received his PhD in Physical Chemistry in 1972, Subsequently, he held positions at the University of Bochum and later on at the University of Hannover. In 1995, he was appointed Professor of Physical Chemistry at Braunschweig Institute of Technology, Braunschweig, Germany. His research activities in physical chemistry of solids put particular emphasis on in-situ solid state spectroscopies applied to defects and diffusion and to the microdynamics and reaction kinetics of solids.

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