

SOFC DEVELOPMENT AT CERAMIC FUEL CELLS LIMITED

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ABSTRACT

Since its incorporation, Ceramic Fuel Cells Ltd. has made significant progress in developing SOFC technology. In this paper, the status of SOFC technology development in CFCL has been reviewed.

KEYWORDS

Solid oxide fuel cells, technology status, CFCL.

REVIEW OF MANUFACTURING PROCESSES FOR FABRICATION OF SOFC COMPONENTS

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ABSTRACT

In order for fuel cell technology to be commercial, it must meet stringent criteria of reliability, lifetime expectations and cost. While materials play an important role in determining these parameters, engineering design and manufacturing processes for fuel cell stack components are equally important. Manufacturing processes must be low cost and suitable for large volume production for the technology to be viable and competitive in the market place. Several processes suitable for the production of ceramic components used in solid oxide fuel cells as well as ceramic coating techniques required for the protection of some metal components have been described.

KEYWORDS

Solid oxide fuel cells, manufacturing processes, ceramic fabrication.

ZrO₂ - Yb₂O₃ - Y₂O₃ SYSTEM: ELECTRICAL CONDUCTIVITY STUDIES AS A FUNCTION OF Yb₂O₃/Y₂O₃ RATIO

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ABSTRACT

The conductivity of Y₂O₃ (or Yb₂O₃) - ZrO₂ and Y₂O₃ - Yb₂O₃ - ZrO₂ with or without incorporation of Al₂O₃, has been investigated as a function of temperature (400 - 1000°C) and time (at 1000°C) with a four - probe DC technique. The contribution of the grain boundary resistivity was determined with impedance spectroscopy. The microstructure and phase assemblage was investigated with XRD, SEM and TEM. The conductivity deterioration with time at 1000°C occurred for all compositions studied. The incorporation of alumina had the effect of reducing the intensity of ageing. The ageing appears to occur due to decomposition of the metastable cubic phase into the equilibrium phase assemblage consisting of dopant-rich cubic phase matrix and tetragonal ZrO₂ precipitates. The activation energy was considerably lower in the high temperature end of the measurement range and was also affected by the ageing process and alumina incorporation in the lattice.

KEYWORDS

Electrolyte, conductivity, ageing, phase assemblage, microstructure.

XRD CHARACTERISATION OF $\text{LaFe}_{1-x}\text{Mg}_x\text{O}_3$ ($x=0-0.2$)

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ABSTRACT

$\text{LaFe}_{1-x}\text{Mg}_x\text{O}_3$ ($x=0-0.2$) was prepared using a reverse-strike coprecipitation method and compacts of the calcined powder were sintered at 1350, 1400 and 1450°C for 6 h. X-ray diffraction patterns of $\text{LaFe}_{1-x}\text{Mg}_x\text{O}_3$ ($x=0-0.2$) were obtained at room temperature. A single orthorhombic phase, space group *Pnma* (group 62) was observed at all Mg dopant levels. Addition of Mg dopant to the material did not appear to dramatically influence the unit cell parameters of LaFeO_3 , however trace secondary phases at all sinter temperatures in the material were noted. A second calcination step is recommended in order to lessen these secondary phases.

KEYWORDS

LaFeO_3 , perovskite, *Pnma*, unit cell volume and orthorhombic.

THE RAMAN SPECTRUM OF LaFeO₃

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ABSTRACT

LaFeO₃ was prepared using a reverse-strike coprecipitation method and compacts of the calcined powder were sintered at 1350, 1400 and 1450°C for 6 h. The Raman spectra of LaFeO₃ were obtained at both low-temperature and room-temperature, with 9 bands observed of a predicted 24 Raman active modes. Mode assignment is determined from comparison with perovskites with the same structure, namely, SmAlO₃ and LaGaO₃ and are as follows: 102 (B_{1g}), ca·140 (B_{2g}), 150 (B_{1g}), 176 (A_g), 227 (B_{3g}), 261 (A_g), 289 (A_g), 413 (A_g), 429 (B_{3g}).

KEYWORDS

Lanthanum ferrite, perovskite, Raman spectroscopy, modes, orthorhombic phase.

REVALUATION OF CATION RADII OF Mn, Co, AND Ni IN SPINEL-TYPE OXIDES

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ABSTRACT

A Revaluation of the Mn, Co, and Ni cation radii in spinel-type oxides was investigated. The starting oxides with the various compositions in the binary systems Mn-Co and Mn-Ni were prepared at 800°C from Mn, Co, and Ni nitrates. The crystals obtained in this experiment were confirmed to be composed of cubic spinel structure. Their precise lattice constants were determined by graphically extrapolating from the plots of measured lattice constants against $\sin^2 \theta$. The values of the oxygen parameter of the spinel-type oxides with various compositions were determined by the Rietveld method. The radii of Mn^{4+} , Mn^{3+} , Mn^{2+} , Co^{3+} , Co^{2+} , and Ni^{2+} cations in tetrahedral and octahedral sites were calculated based on the known cation distributions in spinel-type oxides and the value of an oxygen parameter evaluated in this work. It was found that the cation radii obtained in this study differ significantly from Shannon's values.

KEYWORDS

Cation radii, Mn-Ni spinel-type oxide, Mn-Co spinel-type oxide, Lattice constant, thermistor.

CORROSION BEHAVIOUR OF CERIUM IMPLANTED STAINLESS STEEL AS A POTENTIAL INTERCONNECT MATERIAL FOR SOLID OXIDE FUEL CELLS

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ABSTRACT

This paper presents, results of an investigation on doping of a chromia forming stainless steel with Ce by ion implantation to improve its high temperature oxidation resistance. Ions of Ce were implanted by using a MEVVA ion implanter at an extraction voltage of 40 kV, to doses of 5×10^{15} to 1×10^{17} ions/cm². After implantation samples were oxidised at 800°C for 1h and 24h, or at 900°C for 0.5h in air. Specimens were characterised by SEM-EDX, GAXRD and RBS. Results show that, Ce implantation produces substantial improvement in oxidation behaviour of the alloy.

KEYWORDS

Corrosion, stainless steel, cerium ion implantation, solid oxide fuel cell, interconnect.

EQUILIBRATION KINETICS OF CaTiO₃

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ABSTRACT

Application of the electrical conductivity measurements in monitoring the gas/solid equilibration kinetics for the oxygen/CaTiO₃ system is reported. The equilibration kinetic data were used to determine the chemical diffusion coefficient for CaTiO₃ at 1223 K.

KEYWORDS

Calcium titanate, diffusion, electrical conductivity.

REACTIVITY AT THE OXYGEN/ZIRCONIA INTERFACE

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ABSTRACT

Work function (WF) measurements were applied to determine the electrical effect accompanying oxygen interaction with the surface of yttria-stabilised zirconia (10 mol %) at elevated temperatures (20-1173 K). The obtained WF data indicate that the oxygen/zirconia system reaches equilibrium above 850 K. Then oxidation results in fast oxygen incorporation resulting in the formation of oxygen vacancies. Below 850 K the system is not in equilibrium. Then reactivity of oxygen with zirconia results in the formation of chemisorbed oxygen species with a maximum of chemisorption isobar at 600 K.

KEYWORDS

Zirconia, oxygen chemisorption, reactivity, work function.

SURFACE ELECTRICAL PROPERTIES OF PbZrO₃

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ABSTRACT

In this paper we report the study on surface electrical properties of undoped PbZrO₃ during oxidation and reduction experiments at 500°C using work function measurements. It was observed that the first heating at 500°C leads to a continuous decrease of work function in time while reduction results in more complex of work function vs. time characteristics. Second oxidation results in a rapid initial work function increase, due to changes of oxygen non-stoichiometry, which is followed by a rapid decrease due to structural transition. The work function changes during the third oxidation is determined by changes of oxygen non-stoichiometry.

KEYWORDS

Surface potential, work function, chemisorption, surface layer.

EFFECT OF OXYGEN PARTIAL PRESSURE ON ELECTRICAL CONDUCTIVITY OF CaTiO₃

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ABSTRACT

This paper reports isothermal changes of the electrical conductivity of undoped CaTiO₃ at 1223 K as a function of oxygen partial pressure in the range between 10 and 10⁵ Pa. It was found that the p(O₂) exponent of the electrical conductivity is equal to 1/5.7 and 1/17.6 at high and low oxygen partial pressure, respectively. These values are discussed in terms of the conductivity regimes, involving the p-type and the n-p transition regime.

KEYWORDS

Calcium titanate, electrical conductivity, defect chemistry.

LITHIUM METAL PHOSPHATE CATHODES FOR Li SECONDARY BATTERIES

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ABSTRACT

Two materials, $\text{Li}_{1-x}\text{Ni}_{1-x}\text{M}_x\text{PO}_4$ (M=Fe, Ti, Cr, Co, Sc with $0 \leq x \leq 0.2$), and $\text{Li}_{3-2x}(\text{Fe}_{1-x}\text{M}_x)_2(\text{PO}_4)_3$ (M=Ti, Zr with $0 \leq x \leq 0.2$), were synthesised using standard solid state reaction techniques. The completion of the products and the phase purity was confirmed by powder X-ray diffraction, the conductivity was tested using AC impedance methods, and coin type cells were made to investigate charge/discharge characteristics. Doped $\text{Li}_{1-x}\text{Ni}_{1-x}\text{M}_x\text{PO}_4$ materials did not undergo any phase changes, whereas the $\text{Li}_{3-2x}(\text{Fe}_{1-x}\text{M}_x)_2(\text{PO}_4)_3$ materials were stabilised in the high ionic conducting γ -phase at room temperature. The best room temperature conductivity results shown were from $\text{Li}_{3-2x}(\text{Fe}_{1-x}\text{M}_x)_2(\text{PO}_4)_3$ with M=Ti, and $x=0.05$, an improvement of several orders of magnitude. Discharge capacity was only 60mAh/g for the LiNiPO_4 , and almost non-existent for the $\text{Li}_3\text{Fe}_2\text{PO}_4$, however when doped the capacity increased considerably.

KEYWORDS

Lithium battery, cathode, lithium metal phosphate.

IN SITU ELECTRON MICROSCOPY STUDY OF BaTiO₃ AT ELEVATED TEMPERATURES

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ABSTRACT

This paper reports an *in situ* high resolution transmission electron microscopy study of undoped BaTiO₃. The study was performed at elevated temperatures from 20°C to 1250°C. No grain boundary mobility was observed up to 1250°C. An earlier study indicates that grain boundaries of BaTiO₃ become mobile above 1350°C.

KEYWORDS

Barium titanate, electron microscopy, HRTEM.

INFLUENCE OF OXYGEN PARTIAL PRESSURE ON ELECTRICAL PROPERTIES OF SINTERED OXIDE WITH SPINEL-TYPE STRUCTURE COMPOSED OF Mn, Co, AND Ni

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ABSTRACT

The effect of P_{O_2} atmosphere during annealing on electrical properties of spinel-type oxide composed of Mn, Co, and Ni used as a thermistor material was investigated. Sintered body of monophase spinel-type oxide with composition of $Mn_{1.5}CoNi_{0.5}O_4$ was prepared by oxidation and fired at 1400°C. Oxidation was conducted at 1100°C at which the spinel structure is stable. The spinel-type oxide was annealed in atmospheres of various P_{O_2} in the temperature range 100 to 300°C. The oxide was p-type semiconductor regardless of annealing conditions. The electrical conductivity and the mobility increased with increasing P_{O_2} . Electrical conduction of the oxide was considered to be controlled by a small polaron hopping.

KEYWORDS

Thermistor, spinel, oxygen concentration, electrical conductivity, Seebeck coefficient.

EFFECT OF SURFACE PREPARATION OF ZIRCONIA ON MICROSTRUCTURE AND ITS REACTIVITY WITH OXYGEN

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ABSTRACT

This paper reports surface electrical properties of zirconia, studied using work function measurements in the temperature range 297-1173 K during oxidation. Specifically, the paper considers the effect of surface preparation procedure, such as grinding and polishing, on the reactivity of zirconia with oxygen.

KEYWORDS

Zirconia, surface microstructure, work function, reactivity.

DIFFUSION IN BaTiO₃ UNDER CHEMICAL POTENTIAL GRADIENT

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ABSTRACT

Dc electrical conductivity was applied to monitor isothermal equilibration kinetics for undoped BaTiO₃ single crystal within the p-type regime in the temperature range 950 - 1206 K during both oxidation and reduction experiments. Thus, determined chemical diffusion coefficient can be expressed as:

$$D_{chem} = 4.32 \times 10^{-2} \exp \frac{-79.58 [kJ/mol]}{RT}$$

It was found that the chemical diffusion coefficient exhibits a complex dependence on p(O₂).

KEYWORDS

Barium titanate, chemical diffusion, equilibration, electrical conductivity.

EFFECT OF MINORITY CHARGE CARRIERS ON THERMOPOWER AND ELECTRICAL CONDUCTIVITY BaTiO₃

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ABSTRACT

This paper considers the effect of minority charge carriers (electrons) on the electrical properties, such as (I) the electrical conductivity and thermopower and its dependence on oxygen partial pressure, for p-type BaTiO₃. It is shown that the effect of minority charge carriers on thermopower is substantial even in the p(O₂) range in which the p(O₂) exponent of the electrical conductivity exhibits a constant value.

KEYWORDS

Thermopower, electrical conductivity, barium titanate, defect chemistry.

ELECTRICAL CONDUCTIVITY OF (La,Sr)MnO₃

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ABSTRACT

Defect disorder model for undoped and Sr-doped LaMnO₃ was derived from non-stoichiometry data reported in literature. This model is checked against the electrical conductivity data. The regimes corresponding to oxygen deficit and oxygen excess will be discussed. A good agreement between the random defect model and experimental data of the electrical conductivity was revealed.

KEYWORDS

Electrode materials, oxygen non-stoichiometry, defect chemistry.

ELECTRICAL PROPERTIES OF (La,Sr)MnO₃

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ABSTRACT

This paper presents an overview of the electrical conductivity of the (La,Sr)MnO₃ (LSM), at elevated temperatures and under controlled gas phase composition. Specifically, the effect of oxygen pressure on these properties will be considered in terms of defect chemistry and related charge compensation mechanism.

KEYWORDS

Electrical conductivity, electrode material, defect chemistry.

THE HIGH TEMPERATURE MECHANICAL PROPERTIES OF $\text{La}_{0.8}\text{Sr}_{0.2}\text{Ga}_{1-x}\text{Mg}_x\text{O}_{3-\delta}$

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ABSTRACT

Doped-LaGaO₃ is a possible candidate as an electrolyte in a medium temperature SOFC. This paper examines the high temperature mechanical properties of La_{0.8}Sr_{0.2}Ga_{1-x}Mg_xO_{3-δ} as a function of x (x = 0.02-0.2). The modulus of rupture (MOR) was determined at 800°C to vary with x, although not significantly at values below x = 0.1. The maximum MOR was observed at x = 0.15 to be approximately 110 MPa, which decreased to approximately 90 MPa at x = 0.2. The high temperature fracture toughness was determined to be approximately 1.1 to 1.2 MPam^{1/2} at 800°C, and varied little with x.

KEYWORDS

Doped-LaGaO₃, mechanical properties, microscopy, Raman spectroscopy, thermal expansion.

MIXED CONDUCTIVITY IN Co-DOPED LANTHANUM GALLATE

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ABSTRACT

Materials of the composition $\text{La}_{0.8}\text{Sr}_{0.2}\text{Ga}_{0.85-x}\text{Co}_x\text{Mg}_{0.15}\text{O}_{3-\delta}$ ($x = 0$ to 0.25) were synthesised using standard solid state technique resulting in phase purity and high density. Conductivity measurements at different temperatures and oxygen partial pressures revealed an ionic to metallic-like transition in conduction behaviour with rising Co amount. Samples with low Co contents ($x=0.05$) showed ionic behaviour with an average value of 0.15 S/cm at 900°C while heavy doping ($x=0.25$) resulted in metallic type conduction with a value of 5.43 S/cm at 900°C in air.

KEYWORDS

Lanthanum gallate, synthesis, ionic conductivity, mixed conductivity, oxygen partial pressure dependence.

INVESTIGATION OF ELECTRODE REACTIONS IN SOFC BY GCI AND EIS TECHNIQUES

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ABSTRACT

O₂ reduction and H₂ oxidation reactions in solid oxide fuel cells (SOFC) have been investigated by the galvanostatic current interruption (GCI) and electrochemical impedance spectroscopy (EIS) techniques. For O₂ reduction on Sr doped LaMnO₃ (LSM) cathodes, the apparent reaction parameters evaluated from GCI and EIS measurements are very close. In contrast, for H₂ oxidation reaction on Ni/Y₂O₃-ZrO₂ cermet anodes, the apparent reaction parameters evaluated from GCI are significantly lower than that from EIS. Reaction order of 0.5 and activation energy of ~71 kJmol⁻¹ are obtained from GCI data, whereas a first reaction order and activation energy of ~162 kJmol⁻¹ are obtained from EIS data. The reason for the difference in the overall electrode behavior evaluated from GCI and EIS for H₂ oxidation has been discussed.

KEYWORDS

Electrode reactions, solid oxide fuel cells, current interruption, impedance spectroscopy.

FABRICATION AND PROPERTIES OF LOW TEMPERATURE ELECTROLYTE TUBULAR SOLID OXIDE FUEL CELLS

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ABSTRACT

A general procedure for the extrusion of $\text{Ce}_{0.8}\text{Gd}_{0.2}\text{O}_{1.9}$ tubular electrolytes is detailed. Using a commercial water based binder system, $\text{Ce}_{0.8}\text{Gd}_{0.2}\text{O}_{1.9}$ was extruded, then fired at 1600°C to form high-density electrolyte tubes (98% relative to theoretical). Heat-treatment of the $\text{Ce}_{0.8}\text{Gd}_{0.2}\text{O}_{1.9}$ powder at 900°C for 4 h yielded tubes that were highly dense with a smooth surface finish while non heat-treated powders resulted in an extrusion that was less dense with poor surface finish. Electrodes were applied and then the tubular SOFC was electrically tested. A good open circuit voltage of 0.911 V was achieved for the cell, however, a poor operating current density of 0.0123 Acm^{-2} at 0.7 V was obtained. The results demonstrate the potential of fabricating and operating tubular SOFCs.

KEYWORDS

Cerium oxide, fuel cell, extrusion.

PHASE TRANSITION TEMPERATURES OF $\text{Bi}_8\text{Pb}_5\text{O}_{17}$

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ABSTRACT

The temperatures at which different phases occur in $\text{Bi}_8\text{Pb}_5\text{O}_{17}$ were determined using room temperature and high temperature x-ray diffraction in combination with differential thermal analysis. These temperatures largely depend on the thermal history of the sample examined. When the room temperature β_2 - $\text{Bi}_8\text{Pb}_5\text{O}_{17}$ is re-heated, the relationship of the phases, and their respective temperatures are: below 420°C, pure β_2 - $\text{Bi}_8\text{Pb}_5\text{O}_{17}$; 420-460°C, coexistence of the β_2 -phase and intermediate Φ -phase; 460-590°C, the mixed- Φ -phase; 590-615°C, a mixture of the Φ -phase and the β -phase; above 615°C, single β -phase. On cooling, the high temperature β -phase transfers directly to the β_2 -phase in the temperature range 615-590°C.

KEYWORDS

Bismuth lead oxide, phase transition, powder x-ray diffraction.

FABRICATION/EVALUATION OF THIN WALL ZIRCONIA-YTTRIA TUBES FOR SOLID STATE ELECTROCHEMICAL DEVICES

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ABSTRACT

Zirconia tubes are used in sensors and oxygen separation devices. In this paper a technique for the fabrication of thin and uniform wall thickness tubes has been described. Up to 30 cm long tubes (both open and closed end) with a wall thickness of 350 μ m have been made by dry bag isostatic pressing and sintering. Densities over 99% of the theoretical and microstructure with uniform grain size distribution have been achieved. These tubes when tested in typical device operating environments, for example, under the condition of current flow (for oxygen separation from a gas stream), show excellent long term stability and durability. Life time tests for up to six months of continuous operation have been successfully demonstrated.

KEYWORDS

Zirconia tubes, oxygen separation, ceramic fabrication, sensors, impedance.

ADVANCES IN MANUFACTURING AND OPERATION OF ANODE SUPPORTED SOLID OXIDE FUEL CELLS AND STACKS

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ABSTRACT

In the Solid Oxide Fuel Cell project of the Forschungszentrum Jülich (FZJ), good progress has been made in manufacture, assembly and testing of anode supported planar SOFC stacks. The operating temperature has been successfully reduced to below 800°C. This was achieved by modifications of the anode and cathode microstructures. Cost effective ferritic steel interconnect plates can be used now and work is in progress to develop appropriate contact layers and sealing materials. Reliable stack construction and operation is still an ongoing issue. Further work needs to be invested in long-term testing, internal reforming and temperature cycling.

KEYWORDS

Zirconia, cermet, perovskite, glass, ferritic steel, solid oxide fuel cell.

THE EFFECT OF PROCESSING ON THE AGEING CHARACTERISTICS OF A SOFT PZT

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ABSTRACT

Soft piezoelectric ceramics with high dielectric constants and high d_{33} constants are well suited to hydrophone and actuator applications. To achieve a long-term stable response from a practical device, the ageing rates need to be tightly controlled. This paper discusses various compositional and processing influences on the ageing of several characteristics of TLZ-5 ceramic produced by Thomson Marconi Sonar.

KEYWORDS

Ceramic, lead zirconate titanate, piezoelectric, ageing, processing.

DETERMINATION OF EQUILIBRATION KINETICS OF OXIDE ELECTRODE MATERIALS USING A MANOMETRIC METHOD

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ABSTRACT

The gas/solid equilibration kinetics for electrode oxide materials, such as $(\text{La}_{0.8}\text{Sr}_{0.2})\text{MnO}_3$, using a manometric method, was determined. The reaction kinetics between oxygen and the oxide material was monitored using the measurements of the $p(\text{O}_2)$ changes during isothermic experiments of oxidation and reduction. The procedure of the determination will be described and relevant kinetic equations were derived. The equilibration kinetic data obtained can be used to determine the chemical diffusion coefficient.

KEYWORDS

Oxide electrode, manometric method, diffusion, gas/solid reaction.

EQUILIBRATION KINETICS OF (La,Sr)CoO₃

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ABSTRACT

This paper reports the chemical diffusion coefficient for the electrode material (La_{0.8}Sr_{0.2})CoO₃ in the temperature range 565-1080 K determined using a manometric method. It was found that, in analogy to LSM, the activation energy of the chemical diffusion coefficient during oxidation is approximately twice as large as that during reduction.

KEYWORDS

Oxide electrode, diffusion, manometric method, oxygen transport.

EQUILIBRATION KINETICS OF (La,Sr)FeO₃

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ABSTRACT

The equilibration kinetics and related chemical diffusion coefficient for the electrode material (La_{0.72}Sr_{0.18})FeO₃ was determined using a manometric method in the temperature range 643-1083 K. It was found that oxidation is substantially faster than reduction. A comparative analysis of chemical diffusion data for the series of oxides, such as LSM, LSC and LSF, is presented. This analysis indicates that the oxygen transport is the slowest one for LSM and the fastest one for LSC.

KEYWORDS

Oxide electrode, diffusion, manometric method, equilibration kinetics.

EQUILIBRATION KINETICS OF (La,Sr)MnO₃

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ABSTRACT

The chemical diffusion coefficient for the (La_{0.8}Sr_{0.2})MnO₃ for both oxidation and reduction experiments in the temperature range 850-1070 K was determined using both parabolic and logarithmic equations. Thus obtained diffusion data indicate that the rate of the equilibration process during oxidation exhibits much higher activation energy (155-196 kJ/mol) than that during reduction (117-119 kJ/mol). The discrepancy between the two kinetics is considered in terms of the effect of defect interactions on their mobility.

KEYWORDS

Oxide electrode, diffusion, equilibration, manometric method.

DIFFUSION-CONTROLLED CHEMISORPTION TRANSITIONS FOR (La,Sr)MnO₃

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ABSTRACT

This paper considers the mechanism of oxygen interaction with the surface of oxide electrode materials, such as LSM, and surface related electrical properties. The mechanism involves rapid oxygen chemisorption, resulting in an increase of the surface charge, followed by slow oxygen incorporation, resulting in a shift of chemisorption equilibria and leading, in consequence, to a decrease of the surface charge. The latter process, involving transient chemisorption, is controlled by chemical diffusion. A good agreement between the chemical diffusion coefficient determined from work function and that determined by the manometric method is revealed.

KEYWORDS

Electrode material, chemisorption, diffusion, work function.

EFFECT OF Gd ON FERMI ENERGY OF PbZrO₃

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ABSTRACT

This paper describes the effect of Gd on the Fermi energy of PbZrO₃. The Fermi energy was determined using work function measurements at elevated temperatures and under controlled gas phase composition. It was observed that incorporation of Gd into PbZrO₃ results in the formation of donors. The effect of Gd on the electrical properties of PbZrO₃ will be considered in terms of defect chemistry of this compound.

KEYWORDS

Defect chemistry, Fermi energy, donors, work function, surface potential.

EFFECT OF Gd ON ELECTRICAL CONDUCTIVITY OF CaTiO₃

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ABSTRACT

This paper reports the electrical conductivity of undoped and Gd-doped (15 mol %) CaTiO₃ at elevated temperatures. It was observed that incorporation of Gd into CaTiO₃ results in an increase of the electrical conductivity by about one order of magnitude. The activation energy of the electrical conductivity is 153 kJ/mol for undoped CaTiO₃ and 104 kJ/mol for Gd-doped CaTiO₃. The effect of Gd on electrical properties of CaTiO₃ is discussed in terms of defect chemistry.

KEYWORDS

Calcium titanate, diffusion, electrical conductivity, Gd-doping.

EFFECTS OF OXIDATION AND REDUCTION ON THE WORK FUNCTION AND THE SURFACE CHEMICAL COMPOSITION OF (La,Sr)MnO₃

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ABSTRACT

This paper considers the effect of gas phase composition during high temperature annealing on the electrical properties and surface chemical composition of the electrode material (La,Sr)MnO₃, using work function measurements and X-ray Photoelectron Spectroscopy (XPS). It was observed that the work function (WF) of (La,Sr)MnO₃ increases during oxidation and decreases during reduction reactions in the temperature range of 1023 - 1123 K. Both oxidation and reduction at 1073 K result in an enrichment of the surface layer with La with respect to Mn and Sr. The degree of surface La enrichment in the oxidised sample is slightly higher than that in the reduced sample.

KEYWORDS

(La,Sr)MnO₃, work function, surface composition, oxidation, reduction.

KINETIC REGIMES OF THE GAS/SOLID EQUILIBRATION FOR BaTiO₃

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ABSTRACT

Kinetic regimes describing the equilibration degree of BaTiO₃ as a function of time are considered. When bulk diffusion is rate controlling the gas/solid kinetics then a general diffusion equation is applicable in the entire regime. In this case also two approximate solutions of the diffusion equation are applicable, such as a parabolic approximation and a logarithmic approximation, which are valid at low and high values of the equilibration degree, respectively. Finally, in some cases a surface reaction may be rate controlling the gas/solid reaction. This reaction results in a linear dependence between the equilibration degree and time at the beginning of the equilibration process.

KEYWORDS

Barium titanate, diffusion, equilibration kinetics.

NON-STOICHIOMETRY AND FERMI ENERGY OF (La,Sr)MnO₃

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ABSTRACT

This paper considers the effect of non-stoichiometry in both cation and oxygen sublattices of oxide perovskite-type materials, such as (La,Sr)MnO₃, on the Fermi energy. Specifically, the relationships between the oxygen pressure dependence of non-stoichiometry, on one side, and the oxygen pressure dependence of the Fermi energy, on the other side, is derived. This dependence may be used to evaluate electrical properties from non-stoichiometry data of LSM.

KEYWORDS

Perovskites, Fermi energy, non-stoichiometry.

ELECTRICAL PROPERTIES OF CUBIC ZrO₂ STABILIZED WITH MIX-Ln-OXIDES

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ABSTRACT

A mixture of lanthanide oxides has been applied as a stabilizer of cubic ZrO₂. Preparation of dense material requires sintering above 1723 K for several hours. The material containing 12 mol. % of mixed lanthanide oxides exhibit solid electrolyte properties with the ionic transport number equal to 0.95 above 1100K. The electrical conductivity does not depend on oxygen partial pressure within the pressure range of 10⁻¹⁸ -10⁵ Pa. The determined enthalpy of motion of oxygen vacancies assumes the value of 106.2±0.7 kJ/mole.

KEYWORDS

Zirconia, lanthanide oxides, solid electrolyte, electrical conductivity.

PROPERTIES OF $(\text{La}_{0.7}\text{Sr}_{0.3})_{0.9}\text{Mn}_{1-y}\text{Cr}_y\text{O}_{3-\delta}$ CATHODE MATERIALS FOR SOLID OXIDE FUEL CELL

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ABSTRACT

The starting powders of $(\text{La}_{0.7}\text{Sr}_{0.3})_{0.9}\text{Mn}_{1-y}\text{Cr}_y\text{O}_{3-\delta}$ ($y = 0\sim 0.15$) were synthesized by Glycine-Nitrate-Process (GNP). The powder features and sintering behaviors were characterized. The electrical conductivity of sintered samples has been measured. Using a screen printing technique, the cathode film was coated on YSZ pellets for the measurement of cathodic polarization by means of a current interruption technique. It was found that substituting Cr for Mn in A-site deficient La(Sr)MnO_{3-δ} gave rise to increased sinterability, improved morphological and thermal-chemical stability; moreover, cathode overpotential was reduced without a considerable loss of electrical conductivity.

KEYWORDS

Lanthanum manganites, polarization, cathode, SOFC.

EFFECT OF OXIDATION ON THE SURFACE COMPOSITION OF (La,Sr)CoO₃

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ABSTRACT

This paper reports the effect of oxidation of the electrode material (La,Sr)CoO₃ (LSC) at 1073 K on its surface chemical composition. The study of angular-dependent X-Ray Photoelectron Spectroscopy (XPS) indicates that oxidation at 1073 K results in a significant surface Sr enrichment with respect to Co, and, to a lesser extent, a surface Sr enrichment with respect to La.

KEYWORDS

(La,Sr)CoO₃, surface composition, XPS, oxidation, segregation.

EFFECTS OF LONG TERM ANNEALING ON THE SURFACE CHEMICAL COMPOSITION OF SURFACE-DOPED YTTRIA-STABILISED ZIRCONIA

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ABSTRACT

The effect of long term annealing on the chemical composition within the surface layer of yttria-stabilised zirconia (YSZ), surface doped with Ca, La, Sr, Nb and Ti, was studied. The dopants were introduced by either chemical impregnation (Ca, La, Sr and Nb) or ion-implantation (Ti) of the YSZ surface layer, and subsequent annealing. The incorporation kinetics of these dopants into YSZ were investigated using X-ray Photoelectron Spectroscopy (XPS). The diffusion coefficients of Nb in YSZ at 1200°C were also determined by Secondary Ion Mass Spectrometry (SIMS).

KEYWORDS

YSZ, surface-doped, annealing, surface composition, XPS.

PERFORMANCE OF A METHANE-FUELED SINGLE-CELL SOFC STACK AT VARIOUS LEVELS OF FUEL UTILIZATION

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ABSTRACT

Fuel-gas mixtures representing 10 to 85% utilization of a methane-steam mixture at S/C=2 were fed to a single cell stack with a Ni-based anode at 875°C. Cell voltage and power output were recorded at current densities of 50 to 350 mA/cm². The accompanying anode off-gas composition at some of these conditions were measured using on-line gas chromatography and compared with the compositions predicted by a thermodynamic model based on the assumption of no carbon formation. Electrical losses were measured at a chosen current density at various levels of fuel utilization by the galvanostatic current-interruption technique. Cell voltage stability was monitored for up to ca. 1000h at two levels of fuel utilization. The stack performance was simulated using a mathematical model of the stack; the simulations were compared with the stack test data.

KEYWORDS

SOFC, methane, fuel utilization, stack performance, simulation.

CHARACTERISATION OF CERAMICS BASED ON LITHIUM TITANIUM PHOSPHATE

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ABSTRACT

²⁷Al solid state NMR and x-ray powder diffraction have been used to characterise the effects of different heat treatments and binder phases on Lithium Aluminium Titanium Phosphate, $\text{Li}_{1.3}\text{Al}_{0.3}\text{Ti}_{1.7}(\text{PO}_4)_3$ (LATP). The addition of Li_3PO_4 favours the formation of AlPO_4 and other minor phases at higher temperatures and longer times which decreases the bulk conductivity (σ_b) of LATP by a factor of 2. It appears that the stoichiometry of LATP in the single crystal does not correspond to the batch composition. The presence of Li_2O does not favour formation of the AlPO_4 phase, consistent with higher σ_b and greater thermal stability.

KEYWORDS

Lithium conducting ceramic, x-ray powder diffraction, solid state NMR.

THE EFFECT OF ADDITIVES ON CERAMIC MATERIALS FOR LITHIUM SOLID ELECTROLYTES

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ABSTRACT

Impedance spectroscopy and scanning electron microscopy have been used to characterise the effect of heat treatments on the grain boundary conductivity (σ_{gb}) of Lithium Aluminium Titanium Phosphate (LATP) with various binder phases. LATP has a very low σ_{gb} of 10^{-7} S/cm at room temperature, which hinders its use as a ceramic electrolyte for Li-ion batteries. Using either Li_2O or Li_3PO_4 , this conductivity can be increased by an order of magnitude. We also report for the first time the use of Magnetic Pulse Compaction in the processing of these ceramic materials.

KEYWORDS

Lithium conducting ceramic, binder phases, impedance spectroscopy, SEM.

MEASUREMENT OF PROPERTIES OF SEALANT MATERIALS FOR SOLID OXIDE FUEL CELL SYSTEMS

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ABSTRACT

Thermal expansion of ceramic materials, ceramic cements and steels was studied, and their compatibility with materials used in the SOFC system was evaluated. A number of ceramic cements, a glass paste and a glass ceramic were examined to identify their potential as a sealing material in a tubular fuel cell system. Thermal expansion coefficients of these materials were compared for thermal matching with materials employed in the fuel cell construction, such as yttria stabilised with 8 mole% zirconia (8YSZ) and stainless steel. A reasonable match was found for Macor, a glass ceramic, with one of the steel samples studied and with 8YSZ.

Candidate sealants were tested for bonding quality, which made it evident that the ceramic cements had to be discarded. Good bonds between Macor and one of the steel samples and Macor and 8YSZ were formed, resulting in a gas tight seal. Uncertainty remains regarding the long-term effect of the interface reactions between steel and Macor.

Exposure of the Macor and a steel-Macor-8YSZ assembly to the different fuel cell gas atmospheres revealed that the Macor reacts with hydrogen, the long term effect of which is also unknown.

KEYWORDS

Solid oxide fuel cell, expansion coefficient, glass ceramic, ferritic steel.

MINIMUM ACTIVE LAYER THICKNESS FOR A Ni/YSZ CERMET SOFC ANODE

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ABSTRACT

Nickel/yttria stabilised zirconia (YSZ) cermet anodes for solid oxide fuel cells consisting of 40 vol% nickel and 60 vol% YSZ were fabricated with varying thickness of the active layers. The anode materials were milled to the appropriate particle sizes, air-sprayed onto YSZ substrates and subsequently sintered to produce the anode component. The thicknesses ranged from 3.5 μm to 54 μm in order to find the minimum thickness of active layer for a high performance anode using the least material. The anodes were studied in an atmosphere of hydrogen, water and nitrogen in a 3-electrode setup. Impedance spectroscopy was carried out at a temperature of 1000°C.

It was found that 12 μm was the limiting thickness for a low polarisation resistance. Below a thickness of 12 μm , the polarisation resistance increased in an erratic manner as the reaction zone was reduced. The area specific resistance of the porous nickel current collector was 0.80 Ωcm^2 at 1000°C and 0.05 Ωcm^2 for a 12 μm thick Ni-YSZ-cermet.

KEYWORDS

SOFC, anode, nickel, yttria stabilised zirconia, polarisation resistance.

SURFACE CATION NON-STOICHIOMETRY IN UNDOPED BaTiO₃

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ABSTRACT

This paper considers the effect of high temperature treatment on the local chemistry of the surface region of undoped BaTiO₃. Segregation-induced cation non-stoichiometry has been investigated using X-ray photoelectron spectroscopy (XPS) and secondary ion mass spectrometry (SIMS). Samples were thermally treated at 1000°C in a tube furnace under different oxygen activities, and then cooled to room temperature at different rates. For slowly cooled samples, Ti enrichment is found in the surface region of oxidised BaTiO₃, while less Ti segregation occurs in reduced BaTiO₃. Increasing the cooling rate reduces the degree of Ti segregation, but it does not change the general behaviour of segregation in either oxidised or reduced BaTiO₃.

KEYWORDS

BaTiO₃, surface non-stoichiometry, annealing, XPS, SIMS.

SOLUTION SYNTHESIS AND FABRICATION OF THIN FILMS OF Sr- AND Mg- DOPED LaGaO₃ PEROVSKITES BY PULSED LASER ABLATION

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ABSTRACT

Sr- and Mg- doped LaGaO₃ powders of various compositions (La_{1-x}Sr_xGa_{1-y}Mg_yO_{3-(x+y)/2} where 0.1 ≤ x ≤ 0.2, 0.1 ≤ y ≤ 0.2) were synthesised through a novel solution combustion route. X-ray diffraction patterns indicate a pure primitive-cubic phase for each composition prepared. Thin films of La_{1-x}Sr_xGa_{1-y}Mg_yO_{3-(x+y)/2} of different dopant concentrations were fabricated using a pulsed laser ablation technique. The as-deposited films upon x-ray diffraction analysis were found to be amorphous in nature. The films on annealing at 750°C for 4 h gave cubic single phase materials. The thermal expansion coefficient of the compound La_{0.9}Sr_{0.1}Ga_{0.8}Mg_{0.2}O_{2.85} determined using high-temperature x-ray diffraction was higher than that of yttria-stabilized zirconia.

KEYWORDS

Lanthanum gallate, thin films, laser ablation, solid electrolyte, solid oxide fuel cells.