

Available online at www.sciencedirect.com



Journal of Power Sources 140 (2005) 280-296



www.elsevier.com/locate/jpowsour

# Taxonomies of SOFC material and manufacturing alternatives

Keegan C. Wincewicz, Joyce S. Cooper\*

Design for Environment Laboratory, Department of Mechanical Engineering, University of Washington, Seattle, WA 98195, USA

Received 8 July 2004; accepted 8 August 2004 Available online 8 December 2004

#### Abstract

Material and manufacturing alternatives for solid oxide fuel cells are listed and analyzed. Specifically, four categories of anode materials, five categories of cathode materials, four categories of electrolytes, and three categories of interconnect materials are presented. Design considerations including operating temperatures and compatibilities among stack materials are also highlighted. Similarly, stack manufacturing options are separated into seven categories and developed into process sequences based on the number and type of firing steps. This work is intended to facilitate material and manufacturing assessments through the consideration of the variety of alternatives prior to capital investment for wide-scale production.

© 2004 Elsevier B.V. All rights reserved.

Keywords: SOFC; Design; Materials; Manufacturing; Fabrication

# 1. Introduction

Fuel cell technologies are expected to substantially reduce oil dependency and environmental impacts compared to conventional combustion-based power generation technologies. Solid oxide fuel cells (SOFCs) have the additional advantages of high efficiency, ability to utilize high temperature exhaust for cogeneration or hybrid applications, and the ability for internal reforming. SOFCs are favored for high-power applications and have also been suggested for distributed power and mobile auxiliary power units. As SOFC developers are already making decisions on design and fabrication options for SOFC systems, a study of the materials and manufacturing options follows.

SOFCs have been developed in both planar and tubular design configurations. First, tubular configurations are comprised of circular or flattened tubes connected in series or parallel to form the stack. The flattened tubular design, or the high-power density (HPD) SOFC developed by Siemens-Westinghouse, offers improvements in electronic conductivity and is expected to support automated production [1]. Second, planar designs are comprised of rectangular or circular plates used to facilitate reactant flows and again combined to form stacks.

When compared to tubular stacks, planar stacks are characterized by higher cell power densities [2]. Adler [3] notes that tubular cells have larger electrical resistances due to the longer distance electrons flow (roughly half the inner circumference of the tube) and have mass and heat transfer issues. Tubular stacks, however, have been proven for longer periods of operation (up to 69,000 h for a single tube). Also, although planar are considered more cost-effective to produce than tubular stacks, planar models are still relatively expensive compared to other power sources [3].

Tubular and planar stacks are comprised of five key types of components: an electrolyte, anode, cathode, interconnect, and seals. The electrolyte at the heart of each cell is a doped solid ceramic oxide that facilitates the generation of oxygen vacancies and carries the charge between the half reactions at the cathode and anode. The cathode (or the air electrode) and the anode (or the fuel electrode) are the sites of each half reaction: oxygen is reduced to oxide ions consuming two electrons at the cathode and fuel is reduced forming two electrons at the anode. The interconnect is the electric link to the cathode and protects the electrolyte

<sup>\*</sup> Corresponding author. Tel.: +1 206 543 5040; fax: +1 206 685 8047. *E-mail address:* cooperjs@u.washington.edu (J.S. Cooper).

 $<sup>0378\</sup>text{-}7753/\$$  – see front matter 0 2004 Elsevier B.V. All rights reserved. doi:10.1016/j.jpowsour.2004.08.032

from a reducing atmosphere. The seals, used in some planar designs, bond components together and provide gastightness. Also, whereas the electrolyte, interconnect, and seals are gas tight (directing the flow of reactants), the anode and cathode are porous to enable transport of reactants and products through the components. Also, end plates, current collectors, and other hardware are needed to complete the stack.

An important consideration to both design and manufacturing research for both tubular and planar designs is the reduction of operating temperatures in order to reduce production costs. Specifically, high temperature designs ranging from ~850 to 1000 °C can be reduced to intermediate temperatures in the range of ~750–850 °C or even low temperatures of  $\sim$ 500–750 °C. Lower temperatures allow the use of less expensive and proven metallic interconnects. Lower temperatures also require unproven or very thin electrolytes necessitating special fabrication capabilities, causing a decrease in power density, and limiting internal reforming capabilities. In fact, Ivers-Tiffée et al. [4] suggest temperatures below 600 °C in are not a benefit at all because of minimum reforming temperature requirement.

Works summarizing planar and tubular SOFC design alternatives are provided in textbook format and research works. Specifically, textbooks by Larminie and Dicks [5], EG&G Services [6], Hoogers [7], and most notably Minh and Takahashi [8] and Singhal and Kendall's new release [9] provide summaries of proven and some emerging technologies. Their discussions of SOFC design include descriptions of typical materials use and configurations, the advantages and disadvantages of each design, stack performance relationships, and potential applications issues. More research-oriented discussions provide a review or comparison of typical or novel materials within the context of operating conditions and studies of specific components and can be found in select archival journals and SOFC symposium proceedings. Uncommon is research citing cell and stack performance as a function of materials use, especially in the case of interconnects. The rarest is by far reports of long-term stability of multi-cell stacks.

Summaries of SOFC manufacturing options are provided by Will et al. [10] and Woodward [11]. Specifically, Will et al. provide an analysis of processes based on component thickness and Woodward compares costs for select processes. Also, SOFC manufacturing information for single process-material combinations can also be found again in select archival journals and SOFC symposium proceedings.

This paper presents taxonomies of SOFC materials and manufacturing literature from these and many additional references with an emphasis on material and process alternatives. This work is intended to facilitate material and process selection through the consideration of the variety of design and manufacturing alternatives prior to capital investment for wide-scale production and is part of an environmental life cycle assessment (LCA) of SOFC systems.

#### 2. Taxonomy of SOFC materials

Fig. 1 provides a taxonomy of example SOFC material alternatives. Each class and example material is described as follows for each of the five key stack components.

## 2.1. Electrolyte materials

In an SOFC, the electrolyte is a solid oxide that forms an  $O^{2-}$  charge carrier separating the oxidative and reductive half reactions. In high temperature planar designs, the electrolyte can also function as the support during fabrication. Electrolytes can be categorized as single or bilayer, the latter combining materials to enhance performance. Design requirements for the electrolyte are [3,12–14]:

- ionically conductive (should be characterized by oxygen ion transport numbers close to 1);
- electronically insulating;
- chemically stable at high temperatures;
- chemically stable in reducing and oxidizing environments;
- gas tight/free of porosity;
- production as a uniformly thin layer (to minimize ohmic losses);
- thermal expansion that matches electrodes;
- uses inexpensive materials.

Table 1 provides additional information for the electrolyte materials presented in Fig. 1. Singhal and Kendall [9] note that stabilized zirconia and ceria possessing the fluorite structure has been the most favored SOFC electrolytes with perovskites, brownmillerites, and hexagonal structured oxides as more recent alternatives. Among the candidate materials, zirconia is a relatively cheap base material and is by far the most popular for SOFC electrolyte material. Among the available electrolyte materials, operating temperature is very important to electrolyte performance. This more recently means that low/intermediate temperature planar cells are anode supported because of the electrolyte needs to be comparably thin [1]. At higher temperatures, the electrolyte can be as thick as 150-250 µm because of higher ionic conductivities [1]. Table 2 presents approximate conductivities for select electrolyte materials. Among the materials included and for 600–800 °C, YSB and LSGMC provide the greatest average conductivity. For 800-100 °C, LSGMC and GDC provide the greatest average conductivity.

Badwal and Foger [12] note that with operating temperatures ranging from 800 to  $1000 \,^{\circ}$ C, zirconia has good thermal and mechanical shock resistance when doped with yttria, scandia, samarium, and magnesium as Y<sub>2</sub>O<sub>3</sub>, Sc<sub>2</sub>O<sub>3</sub>, Sm<sub>2</sub>O<sub>3</sub>, and MgO. Among the zirconia electrolyte materials, YSZ is the most used SOFC electrolyte. YSZ is characterized by good chemical and mechanical stability with high quality raw materials available [4]. Most common is approximately 8.5% yittria (called CZP [4,12]). Also, although characterized by lower ion conductivity, 3% yttria (called 3YTZ [4]) has been used because of its higher mechanical stability. Many times



Fig. 1. Taxonomy of example SOFC material alternatives.

5-20% alumina is added to enhance the mechanical properties, the sintered density, and the electrical properties [15]. In spite of its popularity, Ralph et al. [16] suggests it is doubtful YSZ will operate well at temperatures below 700 °C because of decreased ionic conductivity. Also, although it can be produced at thicknesses of 1  $\mu$ m, reliability can be low when the electrolyte is made extremely thin [17].

Another zirconia electrolyte, SSZ, is quite promising and Badwal and Foger [12] note that 8–9% SSZ has been used because of its mechanical stability and an ionic conductivity much higher than YSZ. However, SSZ has issues associated with phase transition, aging, and cost. Specifically, although 8–12% scandia doping has been shown to give optimal oxide conductivity at higher temperatures, scandia has a phase transition around 600–700 °C when doping is over 8%. Also, 7–9% doping has been seen to degrade faster than dopants over 9% [19]. Hirano et al. [20] found that annealing causes a decreased conductivity at 1000 °C and adding Gd<sub>2</sub>O<sub>3</sub>, Y<sub>2</sub>O<sub>3</sub>, CeO<sub>2</sub>, and Al<sub>2</sub>O<sub>3</sub> helps suppress the phase transition. They also found that adding 1% Bi<sub>2</sub>O<sub>3</sub> helped stabilize the SSZ and lowered the sintering temperature. However, Badwal and Foger [12] note that SSZ is costly (due to the high cost of scandium) and because the conductivity has been shown to deteriorate over time.

As an alternative to zirconia, ceria electrolytes have high oxygen conductivity when doped with gadolina, samaria, yittria, and calcium (as GDC, SDC, YDC, and CDC). Although these alternatives have been shown to be more stable than zirconia electrolytes [12], they become unstable at low oxygen partial pressures as well as above 700 °C due to increasing electrical conductivity causing cells to short circuit [5]. Among the four ceria materials, GDC, SDC, and YDC are the most promising and have been shown to outperform CDC, which has not been recently used. Of concern for these materials is reaction with YSZ above 1300 °C with the use of interlayers being an important option despite interfacial resistance issues below 600 °C [21]. Specifically, GDC can be used as a compliment interlayer to YSZ to protect against unfavorable anode and cathode reactions. For example, Tsoga et al. [22] found a 1 µm layer of Ce<sub>0.43</sub>Ar<sub>0.43</sub>Gd<sub>0.1</sub>Y<sub>0.04</sub>O<sub>1.93</sub> helped suppress the diffusion problems using cobalt-containing cathodes and YSZ at sintering temperatures. In this case, the reTable 1 Example electrolyte materials<sup>a</sup>

Acronym	Representative chemical formulas
Zirconia electrolytes	
YSZ	$(ZrO_2)_{1-x}(Y_2O_3)_x (x \sim 0.08 - 0.1)$
SSZ	$(ZrO_2)_x(Sc_2O_3)_{1-x} (x \sim 0.8)$
CaSZ	Zr <sub>0.85</sub> Ca <sub>0.15</sub> O <sub>1.85</sub>
Ceria electrolytes	
GDC	$Ce_xGd_{1-x}O_y$ (x ~ 0.8, y ~ 1.8)
SDC	$Ce_x Sm_{1-x}O_y$ (x ~ 0.8, y ~ 1.9)
YDC	$Ce_x Y_{1-x}O_y$ (x ~ 0.8, y ~ 1.96)
CDC	$Ce_x Ca_{(1-x)}O_y (x \sim 0.9, y \sim 1.8)$
Lanthanum electrolytes	
LSGM	$La_xSr_{1-x}Ga_yMg_{1-y}O_3 \ (x \sim 0.9, y \sim 0.8)$
LSGMC	$La_xSr_{1-x}Ga_yMg_{1-y-z}Co_zO_3$ (x ~ 0.8, y ~ 0.8, z ~ 0.085)
LSGMF	$La_xSr_{1-x}Ga_yMg_{1-y-z}Fe_zO_3$ (x ~ 0.8, y ~ 0.5, z ~ 0.4)
LSGMCF	$La_{0.8}Sr_{0.2}Ga_{0.32}Mg_{0.08}Co_{0.2}Fe_{0.4}O_{3}$
LaAlO <sub>3</sub> -based	$La_{1-x}Ca_xAlO_3$ (x=0.0027–0.008), $La_{1-x}Ba_xAlO_3$ (x=0.1)
Others	
BCY	BaCe <sub>x</sub> Y <sub>1-xy</sub> O <sub>3</sub> ( $x \sim 0.25$ )
YSTh	$(\text{ThO}_2)_{1-x}(Y_2O_3)_x (x \sim 0.08 - 0.1)$
YSHa	$(HfO_2)_{1-x}(Y_2O_3)_x (x \sim 0.08 - 0.1)$
Bismuth oxide-based	$(Bi_2O_3)_x(Nb_2O_5)_{1-x}$ (x ~ 0.25)
Pyrochlorores-based	YZr <sub>2</sub> O <sub>7</sub> , Gd <sub>2</sub> Ti <sub>2</sub> O <sub>7</sub>
Barium brownmillerites	$BaZrO_3$ , $Ba_2In_2O_5$ , $Ba_3In_xAO_y$ (A = Ti, Zr, Ce, Hf), $Ba_3Sc_2ZrO_8$
Strontium brownmillerites	$Sr_2ScAl_xA_yO_z$ (A = Mg, Zn), $Sr_2ScAlO_5$ , $Sr_3In_2HfO_8$

<sup>a</sup> References are [1,2,4,5,9,12–17,19,20,23–34,37–42,76,78,94,104–108].

sistance of a GDC variant interlayer was much less than when the YSZ electrolyte was allowed to react with the cathode. As another option, Balazs and Glass [23] found GDC, SDC, and YDC electrolytes to have the highest conductivity of all the rare-earth oxides (except promethium which was not measured) when doped with cerium oxide because they tend to not react with many other SOFC materials.

Among the four preferred ceria electrolyte materials, GDC has shown higher ionic conductivity than YSZ [13,24,25] with thermal expansion properties nearly identical to ferretic stainless steel interconnects [26] and compatibility with most

cathode materials. However, GDC has issues linked to stability and cost. Specifically, GDC has mixed electronic/ionic conductivity at low oxygen partial pressures and is not as mechanically stable as YSZ [4]. Addition of praseodymium oxide can help the stability. Also, gadolinium is relatively expensive which hampers the economic feasibility of GDC [27].

SDC shows high ionic conductivity for operation below 700 °C [28,29]. For example, Zhu et al. found a high performance of 0.25 W cm<sup>-2</sup> below 400 °C for SDC/carbonate electrolytes [30]. Since SDC is relatively compatible with

Table 2

Approximate and example conductivities (S cm <sup>-1</sup>	) for select electrolyte materials <sup>a</sup>
--	---

	600 °C	700 °C	800 °C	900 °C	1000 °C	Average (600–800 °C)	Rank of average (600–800 °C)	Average (800–1000 °C)	Rank of average (800–1000 °C)	
Zirconia elec	ctrolytes									
YSZ	2.82E-03	8.29E-03	2.00E - 02	4.13E-02	7.64E-02	1.04E - 02	8	4.59E-02	8	
SSZ	2.51E-02	5.38E-02	1.00E-01	1.67E-01	2.58E-01	5.96E-02	5	1.75E-01	5	
CaSZ	1.78E-04	8.69E-04	3.16E-03	9.23E-03	2.28E-02	1.40E-03	9	1.17E-02	9	
Ceria electro	olytes									
GDC	2.82E-02	7.30E-02	1.58E-01	3.01E-01	5.18E-01	8.66E-02	4	3.26E-01	2	
YDC	1.00E - 02	2.01E-02	3.55E-02	5.68E-02	8.46E-02	2.19E-02	6	5.90E-02	6	
CDC	5.01E-03	1.30E-02	2.82E-02	5.36E-02	9.21E-02	1.54E-02	7	5.80E-02	7	
Lanthanum e	electrolytes									
LSGM	3.16E-02	7.69E-02	1.58E-01	2.89E-01	4.79E-01	8.90E-02	3	3.09E-01	4	
LSGMC	5.62E-02	1.20E-01	2.24E-01	3.74E-01	5.77E-01	1.34E-01	2	3.92E-01	1	
Other electro	olytes									
YSB	1.00E-01	1.89E-01	3.16E-01	_	_	2.02E-01	1	3.16E-01	3	
YSTh	5.62E-05	3.12E-04	1.26E-03	4.00E-03	1.06E-02	5.42E-04	10	5.29E-03	10	

<sup>a</sup> Data approximated from [25] and will be dependent upon the electrolyte microstructure, doping, and fabrication/sintering processes.

nickel, many times it is used in combination with SDC/Ni anodes. In fact, Xia et al. [21] demonstrated this design with performances as high as  $397 \text{ mW cm}^{-2}$  at  $600 \,^{\circ}\text{C}$ . Also, SDC has outperformed YDC for similarly prepared cells and temperatures between 500 and  $600 \,^{\circ}\text{C}$  [31].

Finally, YDC-salt electrolytes have been shown to significantly outperform pure YDC electrolytes and YSZ/YDC bilayered electrolytes by a factor of 7 and 3, respectively, at 600 °C [27]. Salts used were NaCl, LiOH, NaOH, and LiCl with weight percentages varying between 10 and 25%. Zhu et al. [27] also note that "pure YDC in intermediate temperatures is not successful due to its poor chemical stability compared to ceria". YDC also has the advantage of being the least expensive electrolyte between YDC, SDC and GDC. Similar to SDC, a YDC electrolyte is well used when paired with a YDC/Ni anode. In fact, Peng et al. [31] used this design and achieved performance as high as  $360 \text{ mW cm}^{-2}$  at  $650 ^{\circ}$ C.

Lanthanum gallate electrolytes<sup>1</sup> provide an alternative to zirconia and ceria materials. The most commonly cited are LSGM and LSGMC. Specifically, several studies have shown LSGM to have an ionic conductivity higher than YSZ [25,32,33] although Yan et al. [34] did note equivalent ionic conductivity to YSZ at 1000 °C. Also, Maric et al. [35] state that an LSGM electrolyte paired with a Ni/SDC anode has the best chance of success at 700–800 °C. This combination was also explored by Inagaki et al. [36] who found comparable performance at 800 °C to a YSZ electrolyte–YSZ/Ni anode–LSM/YSZ cathode combination at 1000 °C.

Problematic issues with LSGM relate to gallium evaporation at low oxygen partial pressures and reducing atmospheres [4,37], long-term mechanical stability with a high creep rate when compared to YSZ [16,34], difficulties in producing thin films [38], and cost [12]. Although LSGM has been demonstrated to perform better than YSZ of equal thickness, LSGM has difficulties being made as thin as YSZ. This means that very thin YSZ can outperform a thick LSGM cell [34] at intermediate/high temperatures noting that Yan et al. [34] were able to make a thin (15  $\mu$ m) LSGM electrolyte using wet processes. Finally, LSGM also can form unwanted second phases at lower temperatures such as SrLaGaO<sub>7</sub> and La<sub>4</sub>Ga<sub>2</sub>O<sub>9</sub> in electrolyte boundaries [26].

Other lanthanum electrolytes include LSGMC, LSGMF, LSGMCF, and LaAlO<sub>3</sub>-based materials. Specifically, the doped lanthanum gallate electrolytes have shown increased performance over LSGM. Ishihara et al. [17] showed that the addition of cobalt (as LSGMC), iron (as LSGMF), and cobalt and iron (as LSGMCF) can increase ionic conductivity such that for the cobalt options, if the cobalt is kept in small amounts, the ionic conductivity can increase without increasing the electrical conductivity. Issues with these electrolytes include cobalt diffusion and excessive thermal expansion (for LSGMCF). Specifically for LSGMC, Ishihara et al. [17] demonstrated compatibility with LSCF cathodes postulating that this was because cobalt diffusion was reduced. In another study, adding magnesium oxide to LSGMC was found to improve the mechanical strength with very little decrease in electrical conductivity [39]. Finally, Singhal and Kendall [9] discuss LaAlO<sub>3</sub> electrolytes as possibly attractive at low and intermediate temperatures despite conductivities lower than YSZ. In addition, Yasuda et al. [40] found the addition of 2 wt.% Al<sub>2</sub>O<sub>3</sub> to LSGM increased the mechanical strength with no effect on ionic conductivity and thermal expansion at 800 °C.

Other electrolytes include BCY, bismuth, thoria, hafnia, and pychlorores options, as well as barium and strontium brownmillerites. Specifically, BCY has demonstrated higher ion conductivity than YSZ below 800 °C and SDC below 600 °C. It has potential at temperatures below 600 °C because it outperformed SDC and YSZ electrolytes with a Ni-O/SDC anode and SSC cathode [41]. Bi<sub>2</sub>O<sub>3</sub> has been stabilized with the addition of metal oxides of yittria, gadolina, and tantalum as Y<sub>2</sub>O<sub>3</sub>, Gd<sub>2</sub>O<sub>3</sub>, Ta<sub>2</sub>O<sub>5</sub>, and others [12]. Although doped bismuth oxides have shown nearly 10 times the ionic conductivity of zirconia, they are not very stable in reducing environments on the cathode side. Although the addition of yttrium to bismuth oxide (BYO) has high ionic conductivity and is stable [42], more research is needed on these materials. Concerning bismuth electrolytes, Badwal and Foger [12] note "that it is highly unlikely that such systems will be used as electrolytes in solid oxide fuel cells without the use of a protective coating of a more inert material such as zirconia". Additionally, electrolytes made from metal oxide stabilized Thoria (as ThO<sub>2</sub>) or stabilized Hafnia (HfO<sub>2</sub>) have shown ionic conductivities much lower than the zirconia options. Finally, Singhal and Kendal [9] discuss pychlorores and brownmillerite options. Pychlorores options including gadolinium titanate (Gd<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub>) and vittrium zirconate (YZr<sub>2</sub>O<sub>7</sub>) are only suitable in limited oxygen partial pressure ranges. Brownmillerites offer high oxide ion conductivities with BaZrO<sub>3</sub> materials doped with yttrium having shown high conductivity and chemical stability but are extremely difficult to process into dense electrolyte layers [32].

As described above, in addition to operating temperature impacts on performance, electrolyte compatibility with other SOFC components is also very important to applicability. Examples are presented in Table 3 for YSZ, LSGMC, LSGMF, and LSGMCF.

#### 2.2. Anode materials

In an SOFC, the anode or the "fuel electrode" is the site where fuel is reduced within each cell. In planar designs, the anode can also function as the support during fabrication. Almost always it is the last layer deposited on tubular designs and is not a support [1]. Anode performances are the least emphasized SOFC component when it comes to

 $<sup>^1</sup>$  Potential lanthanum gallate-based electrolytes for SOFCs not included in Fig. 1 or Table 1 include LGN (LaGa<sub>1-x</sub>Ni<sub>x</sub>O<sub>3</sub>), LS-GMn (La<sub>x</sub>Sr<sub>1-x</sub>Ga<sub>y</sub>Mn<sub>1-y</sub>O<sub>3</sub>), (La<sub>0.9</sub>Nd<sub>0.1</sub>)<sub>0.8</sub>\Sr<sub>0.2</sub>Ga<sub>0.8</sub>Mg<sub>0.2</sub>O<sub>3</sub>, La<sub>x</sub>Sr<sub>1-x</sub>Ga<sub>y</sub>Fe<sub>1-y</sub>O<sub>3m</sub> as summarized by [107].

 Table 3

 Prominent electrolyte material incompatibilities

Electrolyte	Incompatibilities	Forms	Solution	References					
YSZ	LSCF, LSM	SrZrO <sub>3</sub>	Interlayer of GDC, keep below 1200 °C	[1]					
	LaMO <sub>3</sub> -based (M = Mg, Mn, Fe, Co)	$La_2Zr_2O_7$	Interlayer of GDC or SDC, keep below 1200 °C (1000 °C if M=Co)	[37,43–45]					
	Doped ceria above 1300 °C	$Y_{.2}Ce-Y_{.15}Zr$	-Y <sub>15</sub> Zr Keep cell below 1300 °C						
	LnSrO <sub>3</sub> -based (Ln=Pr, Nd, Gd)	Ln <sub>2</sub> Zr <sub>2</sub> O <sub>7</sub> , SrZrO <sub>3</sub>	Sinter at 1000 °C for 100 h	[48,49]					
LSGMC, LSGMF, and LSGMCF	Ni—O	Nickel diffusion	Interlayer of GDC of SDC	[1,34,33,50]					
	Itself or LSM (inconclusive)	SrGaO <sub>3</sub> , La <sub>4</sub> SrO <sub>7</sub> ,	Control Sr/Mg ratios	[37,26]					
		LaSrGaO <sub>4</sub> , LaSrGa <sub>3</sub> O <sub>7</sub> ,	-						
		SrLaGa <sub>3</sub> O <sub>7</sub> , La <sub>4</sub> Ga <sub>2</sub> O <sub>9</sub>							
	LSC	Cobalt diffusion	Protective Interlayer	[51]					

temperature. This is most likely for two reasons: (1) nickel, the most popular anode material, has shown good performance at all temperature ranges, and (2) anode research is orientated towards catalytic breakdown of hydrocarbon fuels.

SOFC anodes are usually made into a cermet to match the thermal expansion of the electrolyte being used to avoid high sintering rates and grain growth/shrinkage [12]. Design requirements for the anode are [3,12–14,33,52–54]:

- electrically conductive;
- high electrocatalytic activity;
- avoid coke deposition;
- large triple phase boundary;
- stable in a reducing environment;
- can be made thin enough to avoid mass transfer losses, but thick enough to provide area and distribute current;
- able to provide mechanical support to electrolyte and cathode if the cell is anode supported;
- thermal expansion coefficient similar neighboring cell component;
- chemically compatible with neighboring cell component;
- has a fine particle size;
- able to provide direct internal reforming (if applicable);
- tolerant to sulfur in fuels (if applicable);
- able to withstand low vapor pressures (will not cause unwanted reactions);
- uses relatively inexpensive materials.

Among the materials listed in Fig. 1, nickel is the most common anode material because it is relatively inexpensive and fulfills most of the anode design requirements. The nickel is usually found in concentrations of 40–60% in the anode cermet to match the thermal expansion of YSZ and must be above the percolation threshold of 30%. To facilitate mass transfer anodes typically have porosities of 20–40% [55].

Despite their popularity, nickel anodes are characterized by a number of problems. Notable are incompatibility with certain electrolytes [26,16] and certain fuels [16,53,56]. Specifically, nickel anodes have been shown to exhibit possible unfavorable reactions with lanthanum electrolytes [26,16,57] as noted in Table 3. In the case of LSGM, protective layers made from CeO<sub>2</sub>, GDC, or SDC [50,1,33,50,58] have been used. Further, nickel anodes are difficult to run on dry methane and higher hydrocarbons because of the formation of carbon fibers above 700 °C [53]. For use of these fuels, there must be sufficient steam for a water gas reaction; for example there must be a steam/methane ratio greater than 2 or 3 [56,53]. Also, whereas Ralph et al. [16] note that the sulfur content of fuels must be below 10 ppm, Singhal and Kendall [9] suggest nickel at high temperatures are sensitive to sulfur concentrations as low as 0.1 ppm. Another issue with nickel anodes is structural damage when thermal cycled repeatedly during stack heating and cooling [59].

Among the nickel anodes, Ni-O/YSZ is the most popular although it has shown reduced thermal expansion mismatches, controlled grain growth, and an increased triple phase boundary area [34]. Ni-O/SSZ cermet anodes have been used by Ukai et al. [60] showing a lower overpotential than Ni-O/YSZ when operating on H<sub>2</sub>/H<sub>2</sub>O fuel at 800 °C and better stability than Ni-O/YSZ. Ni-O/GDC has shown improved electronic and ionic conductivity, catalytic activity, long-term stability, and suppression of carbon formation with methane as a fuel at low steam to carbon ratios [61-64]. Also, Marina et al. [65] found promising results with a 50:50 volume percentage Ni-O/SDC anode to establish good connections at an optimum sintering temperature of 1250–1300 °C [33,65]. Finally, Peng et al. [31] found promising results with Ni-O/YDC anodes utilizing a 65:35 volume percentage at temperatures below 650 °C.

Although alloying copper with nickel has shown reduce carbon formation, the focus of research in alternative anodes has been on replacing the nickel with copper [66]. Copper cermets are less expensive than nickel anodes and have demonstrated better resistance to hydrocarbon coking and YSZ anode cermet densification [53]. Gorte et al. [53], Craciun et al. [56], and Kiratzis et al. [67] describe Cu/CeO<sub>2</sub>/YSZ, Cu/YSZ, and Cu/YZT anodes. Craciun et al. showed that a copper impregnated YSZ anode gave good performance which was increased with the impregnation of CeO<sub>2</sub> because the CeO<sub>2</sub> provided ionic and electronic conductivity [53] and if needed, catalytic activity for hydrocarbon oxidation [68]. Craciun et al. also found the optimal weight percentage of copper was 40% copper which performed nearly equal to nickel. Also, Lu et al. [29] found that a Cu/CeO<sub>2</sub>/SDC anode and a relatively thick SDC electrolyte (at 380  $\mu$ m) ran stable when operating on dry C<sub>4</sub>H<sub>10</sub> between 600 and 700 °C.

For lanthanum anodes, Atkinson et al. [66] suggest  $La_{1-x}Sr_xCrO_3$  as an alternative with good stability and  $La_{1-r}Sr_rCr_{1-v}M_vO_3$  (with M = Mn, Fe, Co, Ni) as an option with improved catalytic properties. Also, Marina et al. [59] found LST (La<sub>0.3</sub>Sr<sub>0.7</sub>TiO<sub>3</sub>) to have good electrical and electrocatalytic properties when sintered in hydrogen at 1650 °C with a thermal expansion similar to YSZ [59]. They also suggest LST will most likely resist structural degradations due to thermal cycling better than a Ni–O/YSZ anode. Sfeir [44] found LAC (LaACrO with A = Sr, Mg, Ca) to inhibit coking but to provide low overall electrocatalytic activity under a pure methane feed. However, the future of perovskite anode materials is likely little because of their high expense and because the anode overpotential is usually small compared to other cell components, money is more likely to be spent on more effective cathode materials.

In addition to nickel, copper, and lanthanum materials, SOFC anodes have been based on ceria, titanium, cobalt, platinum, and ruthenium. Specifically, Marina et al. [65] found good performance with a CeO2/GDC anode, providing  $470 \text{ mW cm}^{-2}$  at  $1000 \,^{\circ}\text{C}$ . Their anode, with 40-50%cerium atoms, is substituted with gadolinium or a similar rare-earth cation and showed a reasonable compromise between conductivities and dimensional stability. Also, a titanium anode, TiO<sub>2</sub>/YSZ, was explored by Mori et al. [69]. Although the TiO<sub>2</sub>/YSZ anode provided an increase in mechanical strength, electrical conductivity decreased with titanium content. The titanium also lowered the firing temperature but decreased the thermal expansion coefficient of the anode cermet. Also, ruthenium anodes described by Ralph et al. [16] demonstrated low overpotential losses and high resistance to carbon deposition and grain growth but are toxic and relatively expensive. Finally, cobalt, iron, and platinum anodes are more expensive and do not show substantial performance improvements over nickel anodes [12].

A recent review article presented by Atkinson et al. [66] describes the implications of anode material choices for SOFCs including a review of fuel related issues and is recommended for further information.

#### 2.3. Cathode materials

The cathode or the "air or oxygen electrode" is the site where oxygen is reduced to oxide ions within each cell. The cathode usually functions as the support during fabrication of tubular cells. In lower temperature SOFCs, the cathode is often the limiting resistance of the SOFC cell because of its large overpotential: usually much larger than anode overpotentials. This is because large activation and concentration polarizations develop at low temperatures [11]. Design requirements for the cathode are [3,13,14,53,54]:

- high electronic conductivity;
- chemically compatible with neighboring cell component (usually the electrolyte);
- can be made thin and porous (thin enough to avoid mass transfer losses, but thick enough to provide area and distribute current);
- stable in an oxidizing environment;
- large triple phase boundary;
- catalyze the dissociation of oxygen;
- high ionic conductivity;
- adhesion to electrolyte surface;
- thermal expansion coefficient similar to other SOFC materials;
- relatively simple fabrication;
- uses relatively inexpensive materials.

Cathode material performance is very dependent on temperature (but less so when compared to the electrolyte), grain size, microstructure, and the formation or deposition process. Table 4 provides additional information for the cathode materials presented in Fig. 1. Among those listed LSM, LSF, and SSC are leading cathode materials. Specifically, LSM and LSF are the proven lanthanum options. First, LSM is the most popular cathode material for high temperature SOFCs because of its stability with zirconia electrolytes. For LSM, the perovskite LaMnO<sub>3</sub> (ABO<sub>3</sub>) is doped both at the A and B sites by cations. Calcium (at 10-30 mol%) and strontium (at 10–20 mol%) are generally doped at the A site [12]. If the strontium concentrations are too low, a decrease has been seen in electric conductivity. Also, LSM is often mixed with YSZ to extend the triple phase boundary, reaction sites and significantly reduce electrode polarization [70]. LSM has also been mixed with platinum to increase oxygen reduction rates [12].

For LSM and other lanthanum-based cathodes, compatibility with YSZ electrolytes is particularly important. Specifically, LSM reacts with YSZ at temperatures above 1300 °C [16,71]. Similarly, YSZ electrolytes are only compatible with LSM if the temperature stays below 1200 °C [37] and the strontium content is below 30% [16]. Yoon et al. [70] saw improved performance with an SDC coating on LSM. For an LSM/YSZ cathode, Hart et al. [71] investigated an LSM/GDC composite layer at the electrolyte-cathode interface which showed higher performance at lower temperatures. As a result of these issues, LSM has been paired with GDC (or other ceria-based interlayers) for lower temperature fuel cells [72].

Despite its lower electrical conductivity [73], LSF is one of the best candidates to replace LSM between 650 and 800 °C [16,74]. Researchers at PNNL and ANNL have focused their interests on LSF cathodes over cobalt (as LSCF), nickel, and manganese B-site cations to improve chemical and longterm stability and power density [47]. In fact, Krumpelt et al. [74] reported lanthanum deficient LSF had significantly lower electrical impedance than stoichiometric LSF

Table 4 Example cathode materials

Acronym	Representative chemical formulas
Lanthanum cath	odes
LSM	$La_x Sr_{(1-x)} MnO_3 \ (x \sim 0.8)$
LSF	$La_x Sr_{(1-x)} FeO_3 \ (x \sim 0.8)$
LSC	$La_x Sr_{(1-x)} CoO_3 \ (x \sim 0.6 - 0.8)$
LSCF	$La_{(1-x)}Sr_xFe_yCo_{(1-y)}O_3 \ (x \sim 0.4, \ y \sim 0.2)$
LSMC	$La_x Sr_{(1-x)} Mn_y Co_{(1-y)} O_3 (x \sim 0.8)$
LSMCr	$(La_xSr_{1-x})_{.91}Mn_yCr_{(1-y)}O_3 (x \sim 0.7, y \sim 0.95)$
LCM	$La_x Ca_{(1-x)} MnO_3 \ (x \sim 0.5)$
LSCu	$La_{(1-x)}Sr_xCuO_{2.5} (x \sim 0.2)$
LSFN	$La_x Sr_{(1-x)} Fe_y Ni_{(1-y)} O_3 (x=0.8, y=0.8)$
LNF	$LaNi_{(1-x)}Fe_xO_3 \ (x \sim 0.4)$
LSCN	$La_x Sr_{(1-x)} Co_y Ni_{(1-y)} O_3$ (x ~ 0.6, y ~ 0.98)
LBC	$La_x Ba_{(1-x)} CoO_3 (x \sim 0.4)$
LNC	$LaNi_{(1-x)}Co_xO_3 \ (x \sim 0.4)$
LSAF	$La_xSr_{(1-x)}Al_yFe_{(1-y)}O_3 \ (x \sim 0.8, y \sim 0.2)$
LSCNCu	$La_x Sr_{(1-x)} Co_y Ni_{(1-y-z)} Cu_z O_3 \ (x \sim 0.8, y \sim 0.8, z \sim 0.05)$
LSFNCu	$La_xSr_{(1-x)}Fe_yNi_{(1-y-z)}Cu_zO_3 \ (x \sim 0.8, y \sim 0.8, z \sim 0.05)$
LNO	LaNiO <sub>3</sub>
Gadolinium cath	odes
GSC	$Gd_xSr_{(1-x)}CoO_3 (x \sim 0.8)$
GSM	$Gd_{(1-x)}Sr_xMnO_3$ (x ~ 0.3–0.6)
Vittria aathodaa	
VSCE	$V_{(1)} = Sr Co E_{2(1)} O_{2} (y = 0.7 row 0.3, 0.8)$
VCCE	$\Gamma_{(1-x)}SI_xCO_y\Gamma_{(1-y)}O_3(y=0.7, x=0.5-0.8)$
VBCu	$\Gamma_{(1-x)} C a_x C O_y \Gamma C_{(1-y)} O_3 (x = 0.2, y > 0.1-0.7)$
I BCu	1 Ba <sub>2</sub> Cu <sub>3</sub> O <sub>7</sub>
Strontium catho	des
SSC	$Sm_xSr_{(1-x)}CoO_3 \ (x \sim 0.5)$
NSC	$Nd_xSr_{(1-x)}CoO_3 \ (x \sim 0.8)$
BSCCu	$Bi_2Sr_2CaCu_2O_8$
Praseodymium o	cathodes
PSM	$Pr_xSr_{(1-x)}MnO_3 (x \sim 0.65)$
PCM	$Pr_xCa_{(1-x)}MnO_3 (x \sim 0.7)$
PBC	$PrxBa_{(1-x)}CoO_3 \ (x \sim 0.5)$
<sup>a</sup> References 90.92,109,1101.	are [1,4,12,13,16–18,24,28,35,37,41,43,47,49,51,68,70–

(i.e., La<sub>0.8</sub>Sr<sub>0.25</sub>FeO<sub>3</sub> performed better than La<sub>0.8</sub>Sr<sub>0.2</sub>FeO<sub>3</sub>). Also, for lower temperature SOFCs, LSF has shown better properties than LSM due to the overpotential of LSM. For example, the LSM overpotential at 1000 °C is 1  $\Omega$  cm<sup>-2</sup> but increases to 2000  $\Omega$  cm<sup>-2</sup> at 500 °C [75]. Already Delphi has used LSF in their power units breaking the trend of LSM commercial cathodes.

The remaining lanthanum cathode materials have shown varying success in conductivity and stability improvements over LSM and LSF. Specifically, LSC is a candidate for lower temperature stacks with a higher conductivity than LSM and one of the better power densities when coupled with an LSGM electrolyte [1,76]. However, some researchers suggest LSC is preferred with ceria electrolytes or with a protective layer of ceria due to large thermal expansion problems and reactivity with zirconia [12,13,77]. In general, LSC's long-term stability is hampered by cobalt diffusion and phase separation (at 750 °C) and has problems with peeling after sintering [1,35,47,51,76].

LSCF is one of the better performers with GDC because of its stability [76] and thermal expansion compatibility [24]. Again, LSCF reacts with zirconia when fired but a protective layer of YDC can help offset this problem [1,78]. Like LSC, LSCF does not react with cerium electrolytes and has a similar thermal expansion coefficient which gives it excellent potential at lower temperatures [76]. Also, LSCF has shown higher activation energies than LSC such that its resistance increases rapidly as temperature decreases; LSCF also has a more compatible thermal expansion coefficient than LSC [16,79].

Improvements have been demonstrated using other lanthanum cathodes but not always without introducing other issues usually in addition to incompatibility with zirconia. Specifically, LSMC has shown increased ion and electrical conductivity over LSM although problems have been demonstrated related to thermal expansion coefficient mismatch with YSZ and the formation of second phases at high cobalt contents [4,12,73]. LSCN has illustrated good performance with GDC [77] although resistance has been found to increase rapidly at temperatures below 800 °C [16] and its long-term stability has been questioned [47]. LSFN is a low temperature candidate found to be stable up to 1400 °C when the iron chemical subscript value is higher than 0.5 [16,80]. Also, Murata and Shimotsu [81] found that when LSMCr was combined with YSZ, performance reached  $1.5 \,\mathrm{W \, cm^{-2}}$ at 1000 °C with hydrogen and oxygen. Chiba et al. [82] explored the use of LNF in the cathode and found a better thermal expansion compatibility with YSZ than that of LSM. They also found LNF to exhibit three times the electronic conductivity as LSM at 800 °C. LNO has shown a relatively low areal resistance on YSZ but relatively high on GDC [16]. Ishihara et al. [18] found LBC to have a comparable performance to SSC at 800 °C and outperform SSC at 600 °C with an LSGM electrolyte despite the formation of BaCO<sub>3</sub> in an atmosphere with greater than 10% CO<sub>2</sub>. LCM has been found stable at high temperatures, has a thermal expansion coefficient compatible with YSZ, and resists La<sub>2</sub>Zr<sub>2</sub>O<sub>7</sub> generation better than LSM [43,83]. Finally and notably, LSCu was found to have no reaction with YSZ with excellent electronic conductivities and small cathode polarization which were more than eight times lower than LSM in similar conditions [68].

No improvements were found related to the use of LNC, LSAF, LSCNCu, and LSFNCu. Specifically, Hrovat et al. [84] found LNC to have a more damaging reaction with YSZ than most perovskites. Also, LSAF has shown a lower electrical ionic and electrical conductivity than LSF [85] and LSCNCu and LSFNCu had relatively large areal resistances on YSZ electrolytes [16]. No performance information was found for LNF and LSCN.

In addition, the use of praseodymium instead of lanthanum in SOFC cathodes has shown decreased cathode overpotentials and enabled higher catalytic activities [86]. Specifically, PCM has demonstrated higher electrical conductivity, lower cathode potential, low-reactivity with YSZ and a more similar thermal expansion to YSZ. PSM was found to react to form  $Pr_2Zr_2O_7$  or SrZrO [87]. PBC has demonstrated potential in low temperature conditions with a BCY electrolyte [41]. Kostogloudis et al. [86] found that of the praseodymium dopants paired with an LSGM electrolyte, cobalt diffused the greatest, followed by iron and then manganese. Another problem in LSGM/praseodymium cathode interface was the formation of an LSGM second phase, LaSrGa<sub>3</sub>O<sub>7</sub> [86].

Strontium cathodes include SSC, NSC, and BSCCu. Specifically, SSC has shown a higher ionic conductivity than LSM, similar performance to LSCF, exchange parameters higher than LSC and LSM [88,89], and to be particularly compatible with GDC and LSGM [17]. A drawback of SSC is that it does not perform as well as GSC and NSC and reacts with YSZ and SSZ (at >900 °C) [88,89]. SSC also has an extremely large areal resistance with YSZ, limiting its applicability at high temperatures [16]. Also, samarium is very expensive and SSC is "not desirable from the cost point of view" [18]. NSC has shown similar resistance properties to GSC with a GDC electrolyte and has potential in lower temperature applications. BSCCu showed good resistance properties on GDC and has potential on lower temperature fuel cells [16].

Gadolina materials include GSC and GSM. GSC is promising for lower temperature SOFCs because it has one of the smallest chances of an unfavorable reaction with GDC [16]. GSC has shown an overpotential at 800 °C nearly one order of magnitude below that of LSM at 1000 °C [49]. It has been shown that as the strontium contents increase, GSC reacts more vigorously with YSZ to form SrZrO<sub>3</sub>. If the strontium content is eliminated, the SrZrO3 reaction is avoided but at temperatures above 1000 °C the formation of Gd<sub>2</sub>Zr<sub>2</sub>O<sub>7</sub> becomes a problem [49]. Despite these issues, Ralph et al. [16] speak of GSC as "a promising new cathode material for use with GDC" because of its very low areal resistance at temperatures as low as 700 °C. A second gadolina cathode is GSM which does not react with GDC but does react with YSZ [73]. The coefficient of thermal expansion better matches YSZ and GDC than that of LSC and increases with increasing Sr contents [73].

Yittria cathodes include YSCF, YCCF, and YBCu. YSCF shows potential in intermediate temperature range (~800 °C) SOFCs with SDC. YSCF has a lower overpotential than LSC (despite an unfavorable reaction with YSZ) but a higher overpotential than LSCF [28]. YCCF has demonstrated better oxygen reduction when compared to LSM although it reacted unfavorably with YSZ [90]. Finally, YBCu has shown low activation energy with potential in low temperature fuel cells, however, no subsequent studies were found [16].

#### 2.4. Interconnect design

In an SOFC, the interconnect is the electric link to the cathode and protects the electrolyte from the reducing reaction. Design requirements for the interconnect are [12,54,91,92]:

- high temperature oxidation and reduction resistance (simultaneous fuel and oxidant gas exposure);
- stable under multiple chemical gas streams;
- very high electrical conductivity;
- high density with "no open porosity";
- strong and high creep resistances for planar configurations;
- good thermal conductivity;
- phase stability under temperature range;
- resistant to sulfur poisoning, oxidation and carburization;
- low materials and fabrication cost;
- matching thermal expansion to other cell components.

Research in both tubular and planar designs that seeks to reduce operating temperatures look to cut interconnect costs. Specifically, the goal is to use cheap and established metallic interconnects below 900 °C instead of the more expensive chromium perovskite materials or metallic interconnects with perovskite-coatings. Perovskites are necessary to give ample oxidation resistance above 900 °C. This is particularly important in planar systems using far more interconnect material per unit of power delivered.

Among the ceramic materials used in SOFCs, doped lanthanum chromate (LaCrO<sub>3</sub>) is the most common option. The doped elements of lanthanum chromate can include cobalt, iron, nickel, magnesium, copper, strontium, calcium, and vanadium [91]. Many times noble metals such as gold, palladium, silver, and platinum are added although these lanthanum chromite composites have shown volatility at temperatures above 800 °C that might be detrimental during long-term operation [1,4]. In general, problems with lanthanum chromate interconnects are related to high cost, sintering difficulties, and warping. The warping issue is related to a tendency to partially reduce at the interface between the fuel gas and interconnect causing the component to warp and the peripheral seal to break [92].

Yang et al. [92] divided metallic alloys into five groups: chromium alloys, ferritic stainless steels, austenitic stainless steel, iron super alloys, and nickel super alloys. Metallic interconnects both with and without coatings have been used. When compared to the use of ceramic materials, metallic interconnects are stronger, easier to form, have higher thermal and electrical conductivities, and negligible ionic conductivities but a thermal expansion coefficient that tends to be higher than most other cell components [37]. Also, ceramics are superior at high temperatures because of oxidation of the metallic interconnects on the cathode side. This oxide layer, usually made of chromium, has a poor conductivity and is "prone to cracking and spalling during long-term operation" [16].

Summaries of interconnect materials have been published by Zhu and Deevi [91,93] and Yang et al. [92]. Both ceramic and metallic materials are discussed in detail and these references are recommended for further information.

#### 2.5. Seal design

In planar SOFCs, seals attach the cell to the interconnect and/or metal frames as well as seal all possible leakage points. Design requirements for the seals are [12,54,95]:

- electrically insulating;
- low cost;
- thermal expansion compatibility with other cell components;
- chemically and physically stable at high temperatures;
- gastight;
- chemically compatible with other components;
- provide high mechanical bonding strength.

Steele et al. [95] divided seals into three categories: rigidbonded seals, compressive seals, and compliant-bonded seals. Specifically, rigid-bonded seals are the most common and are usually made from glass (pyrex) or glass-ceramic materials [16]. Currently the best candidates are SiO<sub>2</sub> glasses [95]. Researchers at Argonne National Laboratory have used

**PROCESS GROUPS** 

Powder preparation for

forming processes
Powder preparation for (1)

deposition onto formed component w/ sintering &

(2) PVD processes

Component formation

without requiring

subsequent firing

Component formation

requiring subsequent firing

Deposition onto formed

component requiring subsequent firing

Heating

SrO–La<sub>2</sub>O<sub>3</sub>–Al<sub>2</sub>O<sub>3</sub>–L<sub>2</sub>O<sub>3</sub>–SiO<sub>2</sub> glasses [96]. Lahl et al. [97] found aluminosilicate base glass sealants with BaO and CaO to be unsuitable for SOFC use and found MgO with varying alumina or TiO<sub>2</sub> content a better possibility. In their evaluation of seals, Stevenson et al. note that glasses can be tailored thermal expansion characteristics, allow hermetic sealing, and are inexpensive and easy to fabricate [95]. However, glasses are brittle, allow only a few compatible options for thermal expansion, and are characterized by detrimental chemical interactions [95].

Compressive and compliant, bonded seals are less common. Compressive seals are generally mica composites with the advantage of being potentially easy to fabricate, usually from the avoidance of the viscous/sealing step. However, there are not a wide variety of these seals and they do not perform well in thermal cycling which can lead to de-coupling of adjacent stack components and possible gas leakage [95]. Finally, Steele et al. suggest no materials in the compliant, bonded category although it is considered an area that does have promise for sealants [95].

SPECIFIC PROCESSES

(applicability: P=planar, T=Tubular, U=unspecified)

(1) Ball milling, filtering, calcining or drying (P,T)

(1) Ball milling, filtering, calcining or drying (P,T)

(1) Chemical Vapor Deposition (CVD) (P,T)(2) Electrochemical Vapor Deposition (EVD) (T)

(2) Triple roll milling, filtering, calcining or drying (P)

(3) Interconnect metal formation and shearing (P)

(2) Wet powder spraying (P)

(4) Interconnect layup (P)

(1) Compaction(2) Extrusion (P,T)

(3) Slip casting (P)

(2) Dip coating (P)

(4) Tape calendering (P)(5) Tape casting (P)

(4) Painting/ pasting (P)

(6) Screen Printing (P)

(7) Slurry Spraying (P,T)
(8) Sol Gel (P)
(9) Spray Pyrolysis (U)
(10) Transfer Printing (U)

(5) Magnitron Sputtering (PVD) (U) (6) RF Sputtering (PVD) (P)

(1) Flame assisted vapor deposition (U)

(3) Electrophoretic Deposition (EPD) (U)

(11) Vacuum Plasma Spraying/ Thermal Spraying (P,T)

(5) Pulsed Laser Deposition (PLD) (U)



(1) Calcining (P)(2) Sintering (P,T)

(3) Drying (P)

Fig. 2. Taxonomy of SOFC material preparation and component fabrication processes.

Process	Elect	rolytes				Cathodes							Anodes					Interconnects			
	YSZ	SSZ	GDC	LSGM	Other electrolyte	LSM	LSM/YSZ	LSF	SSC	LBC	LSC	Other cathode	Ni—O/YSZ	Ni/CeO <sub>2</sub>	CuO <sub>2</sub> /CeO <sub>2</sub> /YSZ	CuO <sub>2</sub> /YSZ	Other anode	Ferretic Steel	Cr-Fe-Y <sub>2</sub> O <sub>3</sub>	Ca-doped LaCrO <sub>3</sub> ceramic	Other interconnect
Ball milling/grinding	х	х				х	х						х						х		
Blanking/slicing, stack calendaring			х	х				х	х				х	х						x	х
Chemical vapor deposition (CVD)	х															х					
Compaction/pressing				х	х	х	x					х	х				х		х		
Dip coating	х											х									
Electrochemical vapor deposition (EVD)	х												х								
Electrophoretic deposition (EPD)	х			х								х									
Extrusion			x			x						х	х								
Flame assisted vapor deposition						x															
Magnitron sputtering (PVD)	х				х																
								х													
Painting/pasting										x			х								
Pulsed laser deposition (PLD)	х																				
RF sputtering (PVD)	х				х												х				
Roll calendering, blanking and slic-																			х		
ing																					
Screen printing	х				х	x	x	х	х			х	х	х			х				
Slip casting	х						x	х					х								
Slurry spraying	х				х	x	x						х				х				х
Sol-gel	х					х															
Spray Pyrolysis	х												х								
Tape calendering	x												х								
Tape casting	x				x	x	x					х	х				x			х	
Triple roll milling						x							х								
Vacuum plasma spraying/thermal spraying	x	x				x	х						x							х	
Wet powder spraving	x																				

# Table 5 Applicability of SOFC manufacturing processes

290



Fig. 3. Non-fired production: no sintering steps.



Fig. 4. Co-fired production: one sintering step, cast layers.

#### 3. Taxonomy of SOFC manufacturing alternatives

For the stack materials described above, a wide variety of possible material preparation and component fabrication options exist. Fig. 2 presents a taxonomy of these SOFC processes for preparation of the electrolyte, anode, and cathode. Specifically, processes are divided into seven classes based on what is accomplished and what preparation is required. The classification was developed from the reviews presented by Will et al. [10] and Woodward [11] which were extensively supplemented by a review of SOFC literature.<sup>2</sup>

Among the processes listed in the taxonomy, some are not as promising as others when commercialization is considered. For example, the use of compaction, electrochemical vapor deposition (EVD), magnitron sputtering (PVD), RF sputtering (PVD), sol–gel, transfer printing, and roll calendering processes require either further development or are not viable [98]. Also, not all processes have been applied to all components and all materials described above. Table 5 summarizes the applicability of the SOFC processes for select materials developed from the review of SOFC literature. Specifically, if the process was used to make a specific component using a specific material, an "x" was placed in the summary table. Although each application was not reviewed on the basis of how well the process produces components in each material or whether or not the processes are viable long term, the summary is intended as an indication of what processes have been used.

To combine the SOFC processes into manufacturing alternatives, the classes of SOFC processes have been developed into sequences dictated by the type and number of firing steps. The results are presented in Figs. 3–8. To create each sequence, manufacturing sequences presented in literature [70,99,100–102] were compared to the classification of SOFC material preparation and component fabrication processes and the applicability assessment presented in Table 5. Also, although not shown, additional scenarios as described by Craciun et al. [56] would replace the sintering steps with calcining steps in the three sintering step multi-fired production scenarios.

#### 4. Discussion

For use in SOFCs, this work identifies four categories of anode materials, five categories of cathode materials, four categories of electrolytes, and three categories of interconnect materials. Electrolyte material considerations are dominated by operating temperature and thickness, anode material considerations by compatibility fuels and thermal expansion mismatches, cathode material considerations by compatibility with electrolytes, and interconnect material considerations by operating temperature. For stack manufacturing, although

<sup>&</sup>lt;sup>2</sup> Data from [10,11,13,14,16,17,20,25,26,28,33,34,39,40,47,49,52–54, 56,70,72,73,81,84,85,89,90–94,99–102,104,105,107–109,111–132].



Fig. 5. Multi-fired production: two sintering steps, anode supported.

many processes have been used and are presented here for a wide variety of manufacturing sequences, further evaluation is needed to determine which prepare components of the desired quality that scale up within the context of mass production for a reasonable cost. Cost considerations will certainly include material and energy (mostly related to firing) costs.

The research described here is part of a larger project analyzing the environmental life cycle of fuel cells for stationary and mobile applications. The project applies life cycle assessment (LCA), a protocol for assessing the environmental aspects (for example, fuel consumption, greenhouse gas emissions, etc.) of a product from various points in their life cycle: from raw materials acquisition through production, use, and disposal [103]. Understanding product design is important to the development of a bill-of-materials for the identification of materials used in the product for the application of LCA. Because the bills-of-materials used in



Fig. 6. Multi-fired production: two sintering steps, electrolyte supported.



Fig. 7. Multi-fired production: three sintering steps, anode supported.



Fig. 8. Multi-fired production: three sintering steps, electrolyte supported.

LCA need to represent equivalent products (for example, a variety of fuel cells capable of moving the same automobile), linking the design alternative above to fuel cell performance is an important research need. Also, understanding the manufacturing alternatives allows the production phase and ultimately materials acquisition to be modeled. For both design and manufacturing, understanding the variety of alternatives that exist prior to wide-scale production facilitates product design and process selection based on environmental criteria prior to large capital investment.

# Acknowledgements

The Ford Motor Company of Dearborn, Michigan, and the Solid State Energy Conversion Alliance (SECA) provided fi-

# References

- F. Tietz, Materials selection for solid oxide fuel cells, Mater. Sci. Forum 426–432 (2003) 4465–4470.
- [2] S.C. Singhal, Solid oxide fuel cells for clean and efficient power generation, Presentation to Boston University, May 30, 2003.
- [3] S. Adler, Solid oxide fuel cells, Course Notes, Department of Chemical Engineering, University of Washington, 2003.
- [4] E. Ivers-Tiffée, A. Weber, D. Herbstritt, Materials and technologies for SOCF-components, J. Eur. Ceram. Soc. 21 (2001) 1805–1811.
- [5] J. Larminie, A. Dicks, Fuel Cell Systems Explained, Wiley, 2000.[6] EG&G Services, Fuel Cell Handbook, 5th ed., 2000.
- http://www.fuelcells.org/fchandbook.pdf. [7] G. Hoogers, Fuel Cell Technology Handbook, CRC Press, New
- York, 2003. [8] N.Q. Minh, T. Takahashi, Science and Technology of Ceramic Fuel
- Cells, Elsevier, 1995. [9] S.C. Singhal, K. Kendall, High-temperature Solid Oxide Fuel Cells:
- Fundamentals, Design and Applications, Elsevier, 2004.
- [10] J. Will, A. Mitterdorfer, C. Kleinlogel, D. Perednis, L.J. Gauckler, Fabrication of thin electrolytes for second-generation solid oxide fuel cells, Solid State Ion. 131 (2000) 79–96.
- [11] H. Woodward, A performance-based, multi-process cost model for solid oxide fuel cells, Master of Science Thesis, Worcester Polytechnic Institute, 2003.
- [12] S.P.S. Badwal, K. Foger, Materials for solid oxide fuel cells, Mater. Forum 21 (1997) 187–224.
- [13] E. Maguire, B. Gharbage, F.M.B. Marques, J.A. Labrincha, Cathode materials for intermediate temperature SOFCs, Solid State Ion. 127 (2000) 329–335.
- [14] S.C. Singhal, Advances in solid oxide fuel cell technology, Solid State Ion. 135 (2000) 305–313.
- [15] T. He, Z. Lu, L. Pei, X. Huang, Z. Liu, W. Su, Electrical properties and applications of (ZrO<sub>2</sub>)<sub>0.92</sub> (Y<sub>2</sub>O<sub>3</sub>)<sub>0.08</sub> electrolyte thin wall tubes prepared by improved slip casting method, J. Alloys Compd. 333 (2003) 231–236.
- [16] J.M. Ralph, J.A. Kilner, B.C.H. Steele, Improving Gd-doped ceria electrolytes for low temperature solid oxide fuel cells, Mater. Res. Soc. Symp. Proc. 575 (2001) 309.
- [17] T. Ishihara, T. Shibayama, M. Honda, H. Nishiguchi, Y. Takita, Transition metal doped LaGaO<sub>3</sub> perovskite fast oxide ion conductor and intermediate temperature solid oxide fuel cell, Mater. Res. Soc. Symp. Proc. 575 (2000) 283.
- [18] T. Ishihara, S. Fukui, H. Nishiguchi, Y. Takita, La-doped BaCoO<sub>3</sub> as a cathode for intermediate temperature solid oxide fuel cells using a LaGaO<sub>3</sub> base electrolyte, J. Electrochem. Soc. 149 (7) (2002) A823–A828.
- [19] S.P.S. Badwal, F.T. Ciacchi, D. Milosevic, Scandia–zirconia electrolytes for intermediate temperature solid oxide fuel cell operation, Solid State Ion. 136–137 (2000) 91–99.
- [20] M. Hirano, O. Takayuki, K. Ukai, Y. Mizutani, Effect of Bi<sub>2</sub>O<sub>3</sub> additives in Sc stabilized zirconia electrolyte on stability of crystal phase and electrolyte properties, Solid State Ion. 158 (2003) 215–223.
- [21] C. Xia, F. Chen, M. Liu, Reduced-temperature solid oxide fuel cells fabricated by screen printing, Electrochem. Solid-State Lett. 4 (5) (2001) A52–A54.

- [22] A. Tsoga, A. Gupta, A. Naoumidis, P. Nikolopoulos, Gadoliniadoped ceria and yttria stabilized zirconia interfaces: regarding their application for SOFC technology, Acta Mater. 48 (2000) 4709–4714.
- [23] G.B. Balazs, R.S. Glass, AC impedance studies of rare earth oxide doped ceria, Solid State Ion. 76 (1995) 155–162.
- [24] V.V. Kharton, F.M. Figueiredo, L. Navarro, E.N. Naumovich, A.V. Kovalevsky, A.A. Yaremchenko, A.P. Viskup, A. Carneiro, F.M.B. Marques, J.R. Frade, Ceria-based materials for solid oxide fuel cells, J. Mater. Sci. 36 (5) (2001) 1105–1117.
- [25] S.C. Sinhal, Solid oxide fuel cells for stationary, mobile, and military applications, Solid State Ion. 152–153 (2002) 405–410.
- [26] B. Steele, A. Heinzel, Materials for fuel-cell technologies, Nature 414 (2001) 345–352.
- [27] B. Zhu, X. Liu, P. Zhou, Z. Zhu, W. Zhu, S. Zhou, Cost-effective yttrium doped ceria-based composite ceramic materials for intermediate temperature solid oxide fuel cell applications, J. Mater. Sci. Lett. 20 (2001) 591–594.
- [28] H. Fukunaga, C. Arai, C. Wen, K. Yamada, Thermal expansion and cathode behaviors of YSCF as SOFC cathode, SOFC VII 449 (2001).
- [29] C. Lu, W.L. Worrell, R.J. Gorte, J.M. Vohs, SOFCs for direct oxidation of hydrocarbon fuels with samaria-doped ceria electrolyte, J. Electrochem. Soc. 150 (2003) A354–A358.
- [30] B. Zhu, X.T. Yang, J. Xu, Z.G. Zhu, S.J. Ji, M.T. Sun, J.C. Sun, Innovative low temperature SOFCs and advanced materials, J. Power Sources 118 (2003) 47–53.
- [31] R. Peng, C. Xia, X. Liu, D. Peng, G. Meng, Intermediatetemperature SOFCs with thin Ce<sub>.8</sub>Y<sub>.2</sub>O<sub>1.9</sub> films prepared by screen printing, Solid State Ion. 152–153 (2002) 561–565.
- [32] S. Haile, Materials for fuel cells, Mater. Today 6 (2003) 24-29.
- [33] X. Zhang, S. Ohara, R. Maric, K. Mukai, T. Fukui, H. Yoshida, M. Nishimura, T. Inagaki, K. Miura, NI-SDC cermet anode for medium-temperature solid oxide fuel cell with lanthanum gallate electrode, J. Power Sources 83 (1999) 170–177.
- [34] J.W. Yan, Z.G. Lu, Y. Jiang, Y.L. Dong, C.Y. Yu, W.Z. Li, Fabrication and testing of a doped lanthanum gallate electrolyte thinfilm solid oxide fuel cell, J. Electrochem. Soc. 149 (9) (2002) A1132–A1135.
- [35] R. Maric, S. Ohara, T. Fukui, H. Yoshida, M. Nishimura, T. Inagaki, K. Miura, Solid oxide fuel cells with doped lanthanum gallate electrolyte and LaSrCoO<sub>3</sub> cathode and Ni-samaria-doped ceria cermet anode, J. Electrochem. Soc. 146 (1999) 2006–2010.
- [36] T. Inagaki, K. Miura, H. Yoshida, R. Maric, S. Ohara, X. Zhang, K. Mukai, T. Fukui, High-performance electrodes for reduced temperature solid oxide fuel cells with doped lanthanum gallate electrolyte. II. La(Sr)CoO<sub>3</sub> cathode, J. Power Sources 86 (2000) 347– 351.
- [37] S.P.S. Badwal, Stability of solid oxide fuel cell components, Solid State Ion. 143 (2001) 39–46.
- [38] K. Murata, H. Okawa, S. Ohara, T. Fukui, Preparation of La(Sr)Ga(Mg)O<sub>3</sub> electrolyte film by tape casting method, SOFC VII 368 (2001).
- [39] J. Akikusa, T. Yamada, K. Adachi, K. Hoshino, Development of low-temperature operation SOFC, SOFC VII 159 (2001).
- [40] I. Yasuda, Y. Baba, T. Ogiwara, H. Yakabe, Y. Matsuzaki, Development of anode-supported SOFC for reduced-temperature operation, SOFC VII 131 (2001).
- [41] T. Hibino, A. Hashimoto, M. Suzuki, M. Sano, A solid oxide fuel cell using Y-doped BaCeO<sub>3</sub> with Pd-loaded FeO anode and Ba<sub>.5</sub>Pr<sub>.5</sub>CoO<sub>3</sub> cathode at low temperatures, J. Electrochem. Soc. 149 (2002) A1503–A1508.
- [42] C. Huang, K. Fung, Degradation of rhombohedral  $(Y_{.25}Bi_{.75})_2O_3$ Solid Electrolyte, SOFC VII 393 (2001).
- [43] W. Schafer, A. Koch, U. Herold-Shmidt, D. Stolten, Materials, interfaces and production techniques for planar solid oxide fuel cells, Solid State Ion. 86–88 (1996) 1235–1239.

- [44] J. Sfeir, LaCrO<sub>3</sub>-based anodes: stability considerations, J. Power Sources 118 (2003) 276–285.
- [45] T. Armstrong, A. Virkar, Performance of solid oxide fuel cells with LSGM-LSM composite cathodes, J. Electrochem. Soc. 149 (12) (2002) A1565–A1571.
- [46] K. Eguchi, N. Akasaka, H. Mitsuyasu, Y. Nonaka, Process of solid state reaction between doped ceria and zirconia, Solid State Ion. 135 (2000) 589–594.
- [47] S.P. Simner, J.F. Bonnett, K.D. Canfield, K.D. Meinhardt, J.P. Shelton, V.L. Sprenkle, J.W. Stevenson, Development of lanthanum ferrite SOFC cathodes, J. Power Sources 113 (2003) 1–10.
- [48] L. Qiu, T. Ichikawa, A. Hirano, N. Imanishi, Y. Takeda,  $Ln_{1-x}Sr_xFe_yO_{3-\delta}$  (Ln=Pr, Nd, Gd; x=.2, .3) for the electrodes of solid oxide fuel cells, Solid State Ion. 158 (2003) 55–65.
- [49] Y. Takeda, H. Ueno, N. Imanishi, O. Yamamoto, N. Sammes, M.B. Phillipps, Gd<sub>1-x</sub>Sr<sub>x</sub>CoO<sub>3</sub> for the electrode of a solid oxide fuel cells, Solid State Ion. 86–88 (1996) 1187–1190.
- [50] K. Huang, J. Goodenough, A solid oxide fuel cell based on Sr and Mg LaGaO<sub>3</sub> electrolyte: the role of a rare earth oxide buffer, J. Alloys Compd. 303–304 (2000) 454–464.
- [51] T. Horita, K. Yamaji, N. Sakai, H. Yokokawa, A. Weber, E. Ivers-Tiffee, Stability at La<sub>0.6</sub>Sr<sub>0.4</sub>CoO<sub>3-d</sub> cathode/La<sub>0.8</sub>Sr<sub>0.2</sub>Ga<sub>0.8</sub>Mg<sub>0.2</sub>O<sub>2.8</sub> electrolyte interface under current flow for solid oxide fuel cells, Solid State Ion. 133 (3) (2000) 143–152.
- [52] J. Morse, R. Graff, P. Hayes, A. Jankowski, Porous thin-film anode materials for solid oxide fuel cells, Mater. Res. Soc. Symp. Proc. 575 (2000) 321–324.
- [53] R. Gorte, S. Park, J. Vohs, C. Want, Anodes for direct oxidation of dry hydrocarbons in a solid oxide fuel cell, Adv. Mater. 12 (2000) 1465–1469.
- [54] P. Singh, J. Stevenson, SECA core technology program overview, in: Proceedings of the Fourth Annual SECA Meeting, April 15, 2003.
- [55] A. Stambouli, E. Traversa, Solid oxide fuel cells (SOFCs): a review of an environmentally clean and efficient source of energy, Renew. Sustain. Energy Rev. 6 (2002) 433–455.
- [56] R. Craciun, S. Park, R.J. Gorte, J.M. Vohs, C. Wang, W.L. Worrell, A novel method for preparing anode cermets for solid oxide fuel cells, J. Electrochem. Soc. 146 (1999) 4019–4022.
- [57] X. Zhang, S. Ohara, H. Okawa, R. Maric, T. Fukui, Interactions of a La<sub>.9</sub>Sr<sub>.1</sub>Ga<sub>.8</sub>Mg<sub>.2</sub>O<sub>3</sub> electrolyte with Fe<sub>2</sub>O<sub>3</sub>, Co<sub>2</sub>O<sub>3</sub> and NiO anode materials, Solid State Ion. 139 (2001) 145–152.
- [58] K. Yamaji, T. Horita, N. Sakai, H. Yokokawa, Comparison between La<sub>0.9</sub>Ba<sub>0.1</sub>Ga<sub>0.8</sub>Mg<sub>0.2</sub>O<sub>2.85</sub> and La<sub>0.9</sub>Sr<sub>0.1</sub>Ga<sub>0.8</sub>Mg<sub>0.2</sub>O<sub>2.85</sub> as SOFCs electrolytes, Solid State Ion. 152–153 (2003) 517– 523.
- [59] O. Marina, N. Canfield, J. Stevenson, Thermal, electrical, and electrocatalytical properties of lanthanum-doped strontium titanate, Solid State Ion. 149 (2003) 21–28.
- [60] K. Ukai, Y. Mizutani, Y. Kume, Current status of SOFC development using scandia doped Zirconia, SOFC VII 375 (2001).
- [61] O. Marina, C. Bagger, S. Primdahl, M. Mogensen, A solid oxide fuel cell with a gadolinia-doped ceria anode: preparation and performance, Solid State Ion. 123 (1999) 199–208.
- [62] M. Joerger, L. Gauckler, Catalytically active anodes for SOFC, SOFC VII 662 (2001).
- [63] S. Livermore, J. Cotton, M. Ormerod, Fuel reforming and electrical performance studies in intermediate temperature ceria-gadoliniabased SOFCs, J. Power Sources 86 (2000) 411–416.
- [64] E. Batawi, Personal connection (e-mail), Sulzer Hexis, 2003.
- [65] O.A. Marina, S. Primdahl, C. Bagger, M. Mogensen, Ceria-based anodes on a YSZ electrolyte: preparation and electrochemical performance, SOFC V 540 (1997).
- [66] A. Atkinson, S. Barnett, R.J. Gorte, J.T.S. Irvine, A.J. Mcevoy, M. Mogensen, S.C. Singhal, J. Vohs, Advanced anodes for hightemperature fuel cells, Nature 3 (2004) 17–27.

- [67] N. Kiratzis, P. Holtappels, C.E. Hartwell, M. Morgensen, J.T.S. Irvine, Preparation and characterization of copper/yittria tatania zirconia cermets for use as possible solid oxide fuel cell anodes, Fuel Cells 1 (2001) 211–218.
- [68] H. Yu, F. Kuan-Zong, La<sub>1-x</sub>Sr<sub>x</sub>CuO<sub>2.5</sub> as new cathode materials for intermediate temperature solid oxide fuel cells, Mater. Res. Bull. 38 (2003) 231–239.
- [69] M. Mori, T. Yamamoto, M. Mori, T. Horita, N. Sakai, H. Yokokawa, M. Dokiya, Evaluation of Ni and Ti-doped Y<sub>2</sub>O<sub>3</sub> stabilized ZrO<sub>2</sub> cermet as an anode in high-temperature solid oxide fuel cells, Solid State Ion. 160 (2003) 1–14.
- [70] S. Yoon, J. Han, S. Nam, T. Lim, I. Oh, S. Hong, Y. Yoo, H. Lim, Performance of anode supported solid oxide fuel cell with La<sub>0.85</sub>Sr<sub>0.15</sub>MnO<sub>3</sub> cathode modified by sol–gel coating technique, J. Power Sources 106 (2002) 160–166.
- [71] N.T. Hart, N.P. Brandon, M.J. Day, N. Lapena-Rey, Functionally graded composite cathodes for solid oxide fuel cells, J. Power Sources 106 (2002) 42–50.
- [72] E.P. Murray, S.A. Barnett, Improved performance in (La, Sr)MnO<sub>3</sub> and (La, Sr)(Co, Fe)O<sub>3</sub> cathodes by the addition of a Gd-doped ceria second phase, SOFC VI 369 (1999).
- [73] H. Sung Yoon, D. Lee, B. Ho Kim, Synthesis and characterization of  $Gd_{1-x}Sr_xMnO_3$  cathode for SOFC, SOFC VI 493 (1999).
- [74] M. Krumpelt, J. Ralph, T. Cruse, Argonne activity overview, in: SECA Core Technology Review, SECA, Sacramento, CA, 2003.
- [75] Doshj, V. Rajiv Richards, J. Carter, X. Wang, M. Krumpelt, Development of solid-oxide fuel cells that operate at 500 °C, J. Electrochem. Soc. 146 (1999) 1273–1278.
- [76] O. Yamamoto, Solid oxide fuel cells: fundamental aspects and prospects, Electrochim. Acta 45 (2000) 2423–2435.
- [77] S. Visco, C. Jacobson, L. De Jonghe, Thin-film Fuel Cells, Lawrence Berkeley National Laboratory, 1997.
- [78] G. Rietveld, P. Nammensma, J.P. Ouweltjes, Status of SOFC component development at ECN, SOFC VII 125 (2001).
- [79] S. Wang, T. Kato, S. Nagata, T. Honda, T. Kaneko, N. Iwashita, M. Dokiya, Performance of a La<sub>0.6</sub>Sr<sub>0.4</sub>Co<sub>0.8</sub>Fe<sub>0.2</sub>O<sub>3</sub>–Ce <sub>0.8</sub>Gd<sub>0.2</sub>O<sub>1.9</sub>–Ag cathode for ceria electrolyte SOFCs, Solid State Ion. 146 (2002) 203–210.
- [80] R. Chiba, F. Yoshimura, Y. Sakurai, Properties of La<sub>1-y</sub>Sr<sub>y</sub>Ni<sub>1-x</sub> Fe<sub>x</sub>O<sub>3</sub> as a cathode material for a low-temperature operating SOFC, Solid State Ion. 152–153 (2002) 575–582.
- [81] K. Murata, M. Shimotsu, Fabrication and evaluation of electrode supported planar SOFC III stack performance, Electrochemistry 8 (2001) 587–591.
- [82] R. Chiba, F. Yoshimura, Y. Sakurai, Investigation of LaNi<sub>1-x</sub>Fe<sub>x</sub>O<sub>3</sub> as a cathode material for solid oxide fuel cells, Solid State Ion. 124 (1999) 281–288.
- [83] H.Y. Tu, M.B. Phillipps, Y. Takeda, T. Ichikawa, N. Imanishi, N.M. Sammes, O. Yamamoto,  $Gd_{1-x}A_xMn_{1-y}Co_yO_{3-\delta}$  (A = Sr, Ca) as a cathode for solid-oxide fuel cells, J. Electrochem. Soc. 145 (1999) 2085–2091.
- [84] M. Hrovat, N. Katsarakis, K. Reichmann, S. Bernik, D. Kuscer, J. Holc, Characterisation of LaNi<sub>1-x</sub>Co<sub>x</sub>O<sub>3</sub> as a possible SOFC cathode material, Solid State Ion. 83 (1996) 99–105.
- [85] G. Coffey, J. Hardy, L. Pedersen, P. Rieke, E. Thomsen, M. Walpole, Electrochemical properties of lanthanum strontium aluminum ferrites for the oxygen reduction reaction, Solid State Ion. 158 (2003) 1–9.
- [86] G. Kostogloudis, C. Ftikos, A. Ahmad-Khanlou, A. Naoumidis, D. Stoever, Chemical compatibility of alternative perovskite oxide SOFC cathodes with doped lanthanum gallate solid electrolyte, Solid State Ion. 134 (2000) 127–138.
- [87] H. Rim, S. Jeung, E. Jung, J. Lee, Characteristics of Pr<sub>1-x</sub>M<sub>x</sub>MnO<sub>3</sub> (M=Ca, Sr) as cathode material in solid oxide fuel cells, Mater. Chem. Phys. 52 (1998) 54–59.

- [88] H. Fukunaga, M. Koyama, N. Takahashi, C. Wen, K. Yamada, Reaction model of dense Sm<sub>.5</sub>Sr<sub>.5</sub>CoO<sub>3</sub> as SOFC cathode, Solid State Ion. 132 (2000) 279–285.
- [89] S. Wang, T. Kato, T. Honda, T. Kaneko, N. Iwashita, A study of low temperature cathodes on SSZ electrolyte, SOFC VII 607 (2001).
- [90] H. Lee, S. Oh, I. Seo, F. Rocholl, H. Wiemhofer, The cathodic activity and interfacial stability of Y<sub>.8</sub>Ca<sub>.2</sub>Co<sub>1-x</sub>Fe<sub>x</sub>/YSZ electrodes, SOFC V 520 (1997).
- [91] W.Z. Zhu, S.C. Deevi, Development of interconnect materials for solid oxide fuel cells, Mater. Sci. Eng. A348 (2003) 227–243.
- [92] Z. Yang, J. Stevenson, P. Singh, Solid oxide fuel cells, Adv. Mater. Process. 161 (6) (2003) 34–37.
- [93] W.Z. Zhu, S.C. Deevi, Opportunity of metallic interconnects for solid oxide fuel cells: a status on contact resistance, Mater. Res. Bull. 38 (2003) 957–972.
- [94] J.W. Sevenson, K. Hasinska, T.R. Armstrong, Electrical and thermal properties of (La, Sr)(Ga, Mg, Co, Fe)O<sub>3</sub>, SOFC VI 275 (1999).
- [95] J. Stevenson, SOFC Seals: Materials Status, SECA Core Technology Program-SOFC Seal Meeting, July 2003.
- [96] M. Krumpelt, ANL Experience and Suggestions for SOFC Seals, SECA Core Technology Program-SOFC Seal Meeting, July 2003.
- [97] N. Lahl, D. Bahadur, K. Singh, L. Singheiser, K. Hilpert, Chemical interactions between aluminosilicate base sealants and the components on the anode side of solid oxide fuel cells, J. Electrochem. Soc. 149 (5) (2002) A607–A614.
- [98] W. Surdoval, Personal communication, 2004.
- [99] N. Hart, N. Brandon, J. Shemilt, Environmental evaluation of thick film ceramic fabrication techniques for solid oxide fuel cells, Mater. Manuf. Process. 15 (1) (2000) 47–64.
- [100] A.D. Little, Assessment of planar solid oxide fuel cell technology, Prepared for the US Department of Energy, 1999. http://www.netl.doe.gov/scng/enduse/refshelf/AdLstack.pdf.
- [101] V. Karakoussis, N.P. Brandon, M. Leach, R. van der Vorst, The environmental impact of manufacturing planar and tubular solid oxide fuel cells, J. Power Sources 101 (2001) 10–26.
- [102] M. Pehnt, Life cycle assessment of fuel cell components, in: V. Wolf, L. Arnold, A.G. Hubert (Eds.), Handbook of Fuel Cells—Fundamentals, Technology and Applications, Wiley, Chichester, 2003.
- [103] R. Heijungs, S. Sangwon, The Computational Structure of Life Cycle Assessment, Kluwer Academic Publishers, Dordrecht, The Netherlands, 2002.
- [104] K. Masuda, A. Kaimai, K. Kawamura, Y. Nigara, T. Kawada, J. Mizusaki, H. Yugami, H. Arashi, Electrochemical reaction kinetics of mixed conducting electrodes on CeO<sub>2</sub>-based solid electrolytes, SOFC V 473 (1997).
- [105] C. Hatchwell, N.M. Sammes, I.W.M. Brown, Fabrication and properties of Ce<sub>.8</sub>Gd<sub>.2</sub>O<sub>1.9</sub> electrolyte-based tubular solid oxide fuel cells, Solid State Ion. 126 (1999) 201–208.
- [106] Y. Du, N.M. Sammes, Fabrication of tubular electrolytes for solid oxide fuel cells using strontium- and magnesium-doped LaGaO<sub>3</sub> materials, J. Eur. Ceram. Soc. 21 (2000) 727–735.
- [107] S. Nessaraj, A. Raj, R. Pattabiraman, Alternate electrolyte material for intermediate temperature solid oxide fuel cells, Transactions of the SAEST 34 (2001) 89–96.
- [108] S.D. Vora, SECA Program at Siemens Westinghouse, Fourth Annual SECA Meeting, April 15, 2003.
- [109] A. Ahmad-Khanlou, F. Tietz, I. Vinke, D. Stover, Electrochemical and microstructural study of SOFC cathodes based on La<sub>.65</sub>Sr<sub>.3</sub>MnO<sub>3</sub> and Pr<sub>.65</sub>Sr<sub>.3</sub>MnO<sub>3</sub>, SOFC VII 476 (2001).
- [110] J. Kilner, S. Skinner, Zero emission power generation using an all perovskite fuel cell, SOFC VII 224 (2001).
- [111] K. Chour, J. Chen, R. Xu, Metal-organic vapor deposition of YSZ electrolyte layers for solid oxide fuel cell applications, Thin Solid Films 304 (1997) 106–112.

- [112] K. Coyne, J. Hammer, S. Laney, F. Pettit, J. Meier, Fundamental Studies for the Durability of Interconnects in Solid Oxide Fuel Cells, SECA Core Technology Workshop, February, 2003.
- [113] J. Zizelman, Development Update on Delphi's Solid Oxide Fuel Cell System: From Gasoline to Electric Power, Fourth Annual SECA Meeting, April 15, 2003.
- [114] T. Fukui, S. Ohara, M. Naito, K. Nogi, Synthesis of NiO-YSZ composite particles for an electrode of solid oxide fuel cells by spray pyrolysis, Powder Technol. 132 (2003) 52–56.
- [115] L. Gavrilova, N. Proskurnina, V. Cherepanov, Phase equilibria in the La–Co–Ni–O system, SOFC VII 458 (2001).
- [116] M. Nguyen, General Electric SECA Solid Oxide Fuel Cell Program., Fourth Annual SECA Meeting, April 15, 2003.
- [117] T. Horita, Y. Xiong, K. Yamaji, N. Sakai, H. Yokokawa, Stability of Fe—Cr alloy interconnects under CH<sub>4</sub>—H<sub>2</sub>O atmosphere for SOFCs, J. Power Sources 118 (2003) 35–43.
- [118] C. Huang, K. Fung, Effect of moisture on the phase stability of Y<sub>2</sub>O<sub>3</sub>-Bi<sub>2</sub>O<sub>3</sub> solid electrolytes, SOFC VI 254 (1999).
- [119] T. Ishihara, S. Fukui, H. Nishiguchi, Y. Takita, Ba(La)CoO<sub>3</sub> cathode for intermediate temperature operating solid oxide fuel cell using LaGaO<sub>3</sub> based electrolyte, SOFC VII 439 (2001).
- [120] V. Karakoussis, M. Leach, R. van der Vorst, D. Hart, J. Lane, P. Pearson, J. Kilner, Environmental emissions of SOFC and SPFC system manufacture and disposal, ETSU Report F/01/00164/REP, 2000.
- [121] N. Kim, B. Ho Kim, D. Lee, Effect of codopant addition on electrical properties of gadolinia-doped ceria electrolyte, SOFC VI 201 (1999).
- [122] S. Krumdieck, O. Sbaizero, A. Bullert, R. Raj, YSZ Layers by pulsed-MOCVD on solid oxide fuel cell electrodes, Surf. Coat. Technol. 167 (2003) 226–233.
- [123] M. Kuroishi, S. Furuya, K. Hiwatashi, K. Omoshiki, A. Ueno, M. Aizawa, Development of tubular type SOFC module by wet process, SOFC VII 88 (2001).
- [124] S. Mukerjee, S. Shaffer, J. Zizelman, L. Chick, S. Baskaran, M. Chou, C. Coyle, J. Deibler, G. Maupin, K. Meinhardt, D. Paxton, T. Peters, V. Sprenkle, S. Weil, R. Williford, Development of a solid oxide fuel cell stack by Delphi and Battelle, SOFC VIII 88 (2003).
- [125] S. Onuma, A. Kaimai, K. Kawamura, Y. Nigara, T. Kawada, J. Mizusaki, Electrochemical oxidation of CH<sub>4</sub> at the interface of Pt electrode and Y<sub>2</sub>O<sub>3</sub> stabilized ZrO<sub>2</sub> electrolyte, SOFC V 483 (1997).
- [126] J.W. Park, Y.K. Lee, Sintering behavior and electrical property of La<sub>.7</sub>Ca<sub>.33</sub>CrO<sub>3</sub> prepared by doctor blade method, SOFC V 1253 (1997).
- [127] A. Pham, B. Chung, J. Haslam, D. Lenz, E. See, R. Glass, Demonstration of high power density planar solid oxide fuel cell stacks, SOFC VII 148 (2001).
- [128] M. Suzuki, H. Sasaki, S. Otoshi, A. Kajimura, M. Ippommatsu, High power density solid oxide electrolyte fuel cells using Ru/Y<sub>2</sub>O<sub>3</sub> stabilized zirconia cermet anodes, Solid State Ion. 62 (1993) 125–130.
- [129] T. Tsai, S.A. Barnett, Effect of LSM-YSZ cathode on thin electrolyte solid oxide fuel cell performance, Solid State Ion. 93 (1997) 207–217.
- [130] V. Ussui, F. Leitao, C. Yamagata, C. Menezes, D. Lazar, J. Paschoal, Synthesis of ZrO<sub>2</sub>-based ceramics for applications in SOFC, Mater. Sci. Forum 416–418 (2003) 681–686.
- [131] Y. Xion, K. Yamaji, N. Sakai, H. Negishi, T. Horita, H. Yokokawa, Electronic conductivity of ZrO<sub>2</sub>—CeO<sub>2</sub>—YO<sub>1.5</sub> solid solutions, SOFC VII 422 (2001).
- [132] Y. Yang, T. Wen, H. Tu, D. Wang, J. Yang, Characteristics of lanthanum strontium chromite prepared by glycine nitrate process, Solid State Ion. 135 (2000) 475–479.