

Review article

## Components manufacturing for solid oxide fuel cells

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### Abstract

A worldwide overview of processing technology of solid oxide fuel cell (SOFC) components is given and the fabrication techniques of ceramic components are summarized for the different types of SOFCs. Generally, a tendency towards up-scalable and automatizable processes is observed. In addition, critical points of interconnect materials and interconnect fabrication are stressed. Especially for planar cell designs, the chromium contamination of the cathode and interfacial corrosion is regarded as the weak points to be solved for demonstration of planar SOFC units.

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### 1. Introduction

Worldwide, several developers of solid oxide fuel cell (SOFC) technology have made significant progress in recent years and are scheduling SOFC systems in the 1–1000-kW range for concept studies, field tests or even as commercial products between 2001 and 2005. This shift of development from laboratory testing to near-market products implies not only a change in operating conditions from “artificial” comparative tests with hydrogen to real conditions using steam-reformed methane, high fuel utilization and long operating periods. It also implies consistently defined material combinations and especially the choice of up-scalable ceramic processing technologies.

Two main issues in SOFC development can be identified as driving forces during recent years: cost

reduction with respect to low-cost materials and simpler processing techniques, and the improvement of durability in long-term operation. In this context, also the aim of decreasing the operating temperature can be understood in terms of longer SOFC operation. Previously, the planar electrolyte-supported SOFCs operated at temperatures around 1000 °C [1,2]. Therefore, either ceramic interconnect materials or rather expensive chromium alloys had to be used. It was not possible to take advantage of much cheaper ferritic steels due to the too high operating temperatures. Reducing the electrolyte thickness to 10–20 µm [3,4], corresponding to a decrease in electrical resistance, enabled the same power output at 150–200 °C lower operating temperatures than for electrolyte-supported cells with an electrolyte thickness of about 200 µm [5]. Also ferritic steels could be applied [6] because operating temperatures remained below the temperature limit of these materials. However, reducing the electrolyte thickness further does not lead to decreased operating temperatures or increased power

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densities, because the contribution of the ohmic resistance to the overall electrical losses is already rather small. Any further decrease in the operating temperature can only be achieved by improving the electrode performance, i.e. reducing the electrode overvoltages especially at the cathode. Hence, this corresponds to a longer component life with respect to lower corrosion of the metallic interconnect. The introduction of fine-grained composite layers of the electrolyte material (yttria-stabilized zirconia, YSZ) and the electrocatalyst (lanthanum manganite for the cathode and nickel metal for the anode) as an inter-layer between the electrolyte and the original electrode has led to remarkable improvements in cell performance. Today power densities of  $0.35 \text{ W/cm}^2$  at  $0.7 \text{ V}$  and  $800 \text{ }^\circ\text{C}$  can easily be achieved [6], a value which was previously obtained under the same experimental conditions at  $900\text{--}1000 \text{ }^\circ\text{C}$ .

The benefit of such improved electrode performance can be used in three ways: (a) directly as

enhanced power output, (b) by lower operating temperature in terms of longer component life and (c) as a buffer for peak electricity demands during operation. This last possibility can be regarded as an option for increased long-term stability because long performance at high loads can lead to deterioration of the fuel cell [7] due to cationic de-mixing [8,9] and subsequent decomposition. Such a phenomenon has already been observed during operation of oxygen permeation membranes [10]. Improved cell performance can also be achieved by optimizing the microstructure of the electrochemically active layers [6,11]. Therefore, improved materials as well as improved processing of the components are the key to obtaining SOFCs with sufficient long-term stability at an acceptable cost level.

As SOFC materials have already been reviewed in the past [12,13], this paper gives a detailed overview of the current SOFC manufacturing techniques applied by actual SOFC developers worldwide.

Table 1  
SOFC typology and currently leading SOFC developers

Solid oxide fuel cells						
Tubular		Planar				
One cell per tube	Several cells per tube	One cell per layer		Several cells per layer		
Ceramic interconnect	Ceramic interconnect	Metallic interconnect		Ceramic interconnect	Metallic interconnect	
SWPC (D/USA)	MHI+EPDC (Nagasaki) (JP)	thick electrolyte Sulzer (CH)	thin electrolyte FZJ (D)	thick electrolyte SOFCo (USA)	thick electrolyte CFCL (AUS)	thin electrolyte Rolls-Royce (GB)
Toto (JP)		ECN (NL)	ECN (NL)	Tokyo Gas (JP)	Sanyo (JP) ( <i>terminated</i> )	
		TMI (USA)	Risø (DK)	MHI+CEPC (Himeji) (JP)	Siemens (D) ( <i>terminated</i> )	
		Ztek (USA)	Global			
		Fuji Electric (JP) ( <i>terminated</i> )	Thermo electric (CAN)	Mitsui (JP)		
		Murata+Osaka Gas (JP) ( <i>terminated</i> )	Allied Signal (USA)	Risø (DK) ( <i>terminated</i> )		
			CFCL (AUS)	Toho Gas (JP)		
				Dornier (D) ( <i>terminated</i> )		

## 2. Cell design

Solid oxide fuel cells can be grouped into tubular and planar designs. Both types may consist of one or several single cells per stacking unit, i.e. on a single tube or in a single layer (Table 1). Furthermore, the planar designs can be divided into stack systems with metallic or ceramic interconnect material as well as into cells with thick (electrolyte-supported) or thin (electrode-supported) membranes with thicknesses usually of 100–250 and 5–20  $\mu\text{m}$ , respectively. Table 1 gives an overview of the diversification of SOFCs together with the main developers involved.

## 3. Ceramic components

Different processes are used for the low-cost fabrication of electrodes and electrolyte for the solid oxide fuel cell. The components produced must display specific properties to ensure cyclable and long-term stable operation in the fuel cell stack.

The fabrication routes for the individual cell components of the different SOFC designs (planar or tubular) differ greatly depending on which cell com-

ponent is to perform the supporting function in the cell. Whereas for the planar concept in most cases the electrolyte or the anode ensures the mechanical stability of the cells (Tables 2 and 3), in the tubular concepts the cathode or an inert tube is the supporting component (Table 4). The SOFC developers of tubular cells have been looking for alternative, cost-effective coating techniques for electrolyte membrane and anode deposition for more than 5 years to replace the expensive and complicated electrochemical vapor deposition. Wet ceramic techniques are favored due to cost aspects and such work is under way.

In the case of electrolyte-supported cells (Table 2), the fabrication of the electrolyte and of the electrodes is dominated by tape casting and screen printing, respectively. Both fabrication processes are well-established methods in the electroceramics industry and a scale-up is easily feasible. The thickness of the components varies only little: the membrane foils have a thickness between 100 and 200  $\mu\text{m}$ , and both the cathode and the anode are screen printed onto the electrolyte sheets with thicknesses of 40–60  $\mu\text{m}$ . The tape cast electrolyte foils usually have a size of up to  $10 \times 10$  cm, because larger tapes are difficult to handle after sintering.

Table 2  
Developers of SOFC in electrolyte-supported planar cell design and corresponding fabrication and design details

Company	Country	Component	Material	Production process	Thickness	References
Sulzer Hexis	CH	Electrolyte	YSZ	Tape casting	ns	[14]
		Cathode	(La,Sr)MnO <sub>3</sub>	Screen printing	ns	
		Anode	Ni/YSZ	Screen printing	ns	
ECN/InDec	NL	Electrolyte	YSZ	Tape casting	ns	[15,16]
		Cathode	(La,Sr)MnO <sub>3</sub>	Screen printing	50 $\mu\text{m}$ (two layers)	
		Anode	Ni/YSZ	Screen printing	graded composite	
Fraunhofer Ges., IKTS	D	Electrolyte	YSZ	Tape casting	150 $\mu\text{m}$	[17]
		Cathode	(La,Sr)MnO <sub>3</sub>	Screen printing	ns, two layers	
		Anode	Ni/YSZ	Screen printing	ns	
CFCL	AUS	Electrolyte	3YSZ, 8YSZ	Tape casting	100 $\mu\text{m}$	[18,19]
		Cathode	(La,Sr)MnO <sub>3</sub>	Screen printing	50–60 $\mu\text{m}$	
		Anode	Ni/YSZ	Screen printing	50 $\mu\text{m}$	
SOFCo	USA	Electrolyte	YSZ, (Ce,Sm)O <sub>2</sub>	Pressing and sintering	180 $\mu\text{m}$ , 300 $\mu\text{m}$	[20,21]
		Cathode	(La,Sr)CoO <sub>3</sub>	Screen printing	ns	
		Anode	Ni/YSZ	Screen printing	ns	
Tokyo Gas	JP	Electrolyte	3YSZ	Tape casting	50–100 $\mu\text{m}$	[22]
		Cathode	(La,Sr)MnO <sub>3</sub>	Screen printing	150 $\mu\text{m}$	
		Anode	Ni/(Ce,Y)SZ	Screen printing	30 $\mu\text{m}$	
Mitsui Eng. & Shipbuilding	JP	Electrolyte	8YSZ	Tape casting	300 $\mu\text{m}$	[23,24]
		Anode	Ni/YSZ	Painting	150 $\mu\text{m}$	
		Cathode	(La,Sr)(Mn,Cr)O <sub>3</sub>	Painting	150 $\mu\text{m}$	

ns=not specified.

Other developers aiming at the kW range during the next few years prefer planar anode-supported SOFCs due to their potential of lowering the operating temperature. Here several companies and research organizations in Australia, America and Europe have concentrated on cells with a thick, porous anode substrate and a 5–20- $\mu\text{m}$ -thin electrolyte membrane. Besides the frequently used ceramic processing techniques, i.e. tape casting and screen printing, also alternative methods like warm pressing, tape calendaring and wet powder spraying are under investigation. The main selection criteria for the future fabrication route are the cost aspects, the potential for

automation, reproducibility and precision of the different techniques.

In the anode substrate concept the anode is the supporting component of the cell and must therefore display sufficient mechanical stability. The substrates are predominantly produced by tape casting. Pressing processes are very rarely applied and extrusion molding as for the supporting tubes not at all (Table 4). The reason is presumably that the substrates in such processing routes can hardly be fabricated thinner than 1 or 1.5 mm and most of the developers aim at substrates with thicknesses of around 0.5 mm. Usually an anode functional layer of a few micrometers in thickness is

Table 3  
Developers of SOFC in anode-supported planar cell design and corresponding fabrication and design details

Company	Country	Component	Material	Production process	Thickness	References
Sulzer Hexis	CH	Anode substrate	Ni/YSZ	Tape casting	250–500 $\mu\text{m}$	[25]
		Electrolyte	YSZ/(Ce,Y)O <sub>2</sub>	Reactive magnetron sputtering	5/1 $\mu\text{m}$	[26]
ECN/InDec	NL	Cathode	La <sub>0.6</sub> Sr <sub>0.4</sub> Co <sub>0.2</sub> Fe <sub>0.8</sub> O <sub>3</sub>	Screen printing	ns	[26]
		Anode substrate	Ni/YSZ	Tape casting	500–800 $\mu\text{m}$	[15,16,27]
		Anode	Ni/YSZ	Screen printing	3–7 $\mu\text{m}$	[27]
		Electrolyte	YSZ	Screen printing	7–10 $\mu\text{m}$	[28]
FZJ	D	Cathode	(La,Sr)MnO <sub>3</sub> +YSZ	Screen printing	ns	[27]
		Anode substrate	Ni/YSZ	Tape casting	200–500 $\mu\text{m}$	[29]
		Anode substrate	Ni/YSZ	Warm pressing	1500 $\mu\text{m}$	[6,30]
		Anode	Ni/YSZ	Vacuum slip casting	5–15 $\mu\text{m}$	[6,30]
		Electrolyte	YSZ	Vacuum slip casting	5–30 $\mu\text{m}$	[6]
		Electrolyte	YSZ	Reactive magnetron sputtering	2–10 $\mu\text{m}$	[31]
Risø	DK	Cathode	(La,Sr)MnO <sub>3</sub> +YSZ	Wet powder spraying	50 $\mu\text{m}$	[6,32]
		Anode substrate	Ni/YSZ	Tape casting	200–300 $\mu\text{m}$	[33]
		Electrolyte	YSZ	Wet powder spraying	10–25 $\mu\text{m}$	[34]
		Cathode	(La,Sr)MnO <sub>3</sub> +YSZ	Screen printing	50 $\mu\text{m}$	[34]
Global Thermoelectric	CAN	Anode substrate	Ni/YSZ	Tape casting	1000 $\mu\text{m}$	[35]
		Electrolyte	YSZ	Vacuum slip casting	10 $\mu\text{m}$	[35]
		Electrolyte	YSZ	Screen printing	ns	[36]
		Cathode	(La,Sr)MnO <sub>3</sub>	Screen printing	40 $\mu\text{m}$	[35]
Allied Signal	USA	Anode	Ni/YSZ	Tape casting and calendaring	100 $\mu\text{m}$	[37,38]
		Electrolyte	YSZ	Tape calendaring	5–10 $\mu\text{m}$	[37,39]
		Cathode	Doped LaMnO <sub>3</sub>	Tape calendaring	ns	[39]
CFCL	AUS	Anode substrate	Ni/YSZ	Tape casting	500–700 $\mu\text{m}$	[18,19,40]
		Electrolyte	YSZ	Lamination and sintering	10–30 $\mu\text{m}$	[19,40]
		Electrolyte	YSZ	Reactive magnetron sputtering	<16 $\mu\text{m}$	[41]
		Cathode	(La,Sr)MnO <sub>3</sub>	Screen printing	ns	[19,40]
Mitsui Eng. and Shipbuilding	JP	Anode substrate	Ni/YSZ	ns	1000 $\mu\text{m}$	[23]
		Electrolyte	8YSZ	ns	30 $\mu\text{m}$	[23]
		Cathode	(La,Sr)(Mn,Cr)O <sub>3</sub>	ns	150 $\mu\text{m}$	[23]

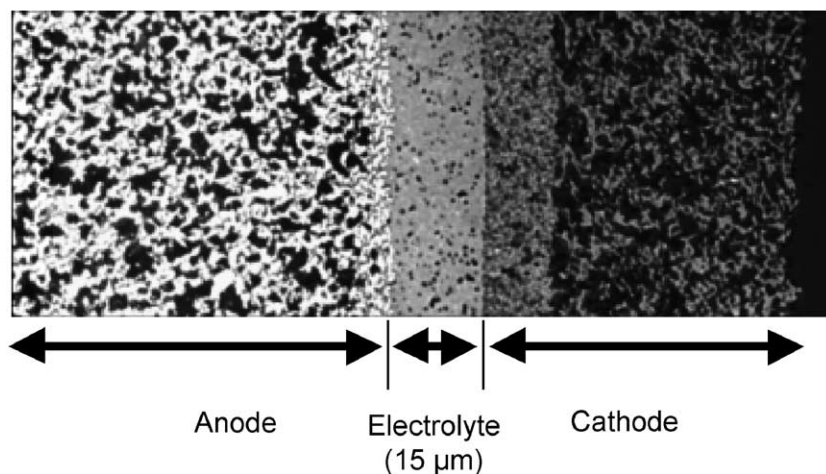


Fig. 1. Microstructure of an anode-supported SOFC with a 15- $\mu\text{m}$ -thick solid electrolyte. Please note that the anode substrate is about 30–100 times thicker than the electrolyte layer (see Table 3).

then deposited onto the substrate to enhance the electrochemical performance [6]. A widely used deposition technique for the thin anode, electrolyte and cathode layers is screen printing (Table 3). A cross-

section through the electrochemical active layers of an anode-supported cell is shown in Fig. 1. In few cases slip casting and wet powder spraying is applied, whereas magnetron sputtering is a curiosity for achiev-

Table 4

Developers of SOFC in tubular cell design and corresponding fabrication and design details

Company	Country	Component	Material	Production process	Thickness	References
SWPC	USA	Cathode tube	Doped LaMnO <sub>3</sub>	Extrusion and sintering	2200 $\mu\text{m}$	[42]
		Electrolyte	YSZ	Electrochemical vapor deposition (EVD)	40 $\mu\text{m}$	[42]
		Anode	Ni/YSZ	Slurry coating or EVD	100 $\mu\text{m}$	[42]
Toto+KEPC	JP	Interconnect	Doped LaCrO <sub>3</sub>	Plasma spraying	85 $\mu\text{m}$	[42]
		Cathode tube	(La,Sr)MnO <sub>3</sub>	Extrusion and sintering	ns	[43,44]
		Electrolyte	YSZ	Slurry coating	40 $\mu\text{m}$	[44]
MHI+EPDC	JP	Anode	Ni/YSZ	Slurry coating	thick film	[43,44]
		Interconnect	(La,Ca)CrO <sub>3</sub>	Slurry coating	ns	[43]
		Substrate tube	Ca-SZ	Extrusion	$\varnothing$ (out) 21 mm	[45–47]
		Cathode	LaCoO <sub>3</sub>	APS	150–200 $\mu\text{m}$	[46]
		Electrolyte	new: (La,Sr)MnO <sub>3</sub>	new: slurry coating		[48]
Rolls-Royce	GB	Electrolyte	YSZ	Low pressure plasma spraying	100–150 $\mu\text{m}$	[47]
		Anode	Ni/YSZ	new: slurry coating		[48]
		APS	80–100 $\mu\text{m}$	[46,47]		
		new: slurry coating		[48]		
		Interconnect	NiAl/Al <sub>2</sub> O <sub>3</sub>	APS	80–100 $\mu\text{m}$	[46,47]
new: (Ln,AE)TiO <sub>3</sub> <sup>a</sup>	new: slurry coating		[48]			
Rolls-Royce	GB	Substrate tube	ns	ns	ns	
		Anode	ns	Printing	ns	[49]
		Electrolyte	ns	Wet slurry printing	<20 $\mu\text{m}$	[49]
		Cathode	ns	Printing	ns	[49]

<sup>a</sup> Ln=lanthanide element, AE=alkaline earth element.

ing very thin layers rather than a realistic approach for cost-effective products with high throughput. Also rather seldom is plasma spraying either as vacuum plasma spraying (VPS) for anodes and electrolytes [50] or as atmospheric (APS) or flame spraying (FS) for electrolytes and cathodes [51]. However, the cost targets of commercial SOFC are difficult to achieve with plasma spraying techniques. Therefore these techniques, commonly applied for tubular systems in the past, are being increasingly replaced by slurry processing (see Table 4). In the tubular fuel cell system of Siemens Westinghouse or Toto the cathode is the supporting cell component. The cathode tubes are produced using an extrusion process with subsequent sintering and the other components are fabricated by slurry coating (Toto) or by plasma spraying and electrochemical vapor deposition (Siemens Westinghouse). Similarly, Mitsubishi Heavy Industries (MHI) also now applies slurry coatings but the supporting tubes are made of stabilized zirconia. Finally, Rolls-Royce's concept is also based on an inert spinel-type rectangular tube on which the fuel cell components are deposited by screen printing.

Apart from tape casting, calendering, screen printing, slip casting, plasma spraying, wet powder spraying, electrochemical vapor deposition for the manufacturing of electrolytes or the coating of substrates with electrolyte layers, other methods have been applied and tested such as laser ablation, multiple spin coating, colloidal deposition, reactive magnetron sputtering, chemical vapor deposition, spray pyrolysis and electrophoresis. All these alternative methods are of scientific interest rather than having the potential to be commercially relevant. A brief overview is also provided by Minh [52].

#### 4. Interconnect materials

The interconnect in SOFCs is the component which electrically connects the single cells and in planar systems additionally separates the gas compartments. In an SOFC system, a number of demands are made on the interconnect, which ultimately determine the material selection. Important requirements are good electrical conductivity, gastightness, chemical compatibility with the adjacent components of the fuel cell, chemical stability in reducing and oxidizing atmos-

pheres, matched thermal expansion and last but not least reasonable costs. In order to meet these requirements, two classes of materials are commonly used for the interconnect, namely, ceramic and metallic materials. Whereas ceramic interconnects played a dominant role in the early SOFC developments and are still essential in tubular designs, metallic interconnects have been frequently used in recent developments. Both variants have benefits and disadvantages and the final choice is therefore always a compromise depending, among other aspects, on the design, the operating temperature, the required service life as well as on the material and production costs of these components.

Practically all the ceramic interconnects of present SOFC systems are based on the perovskite structure of the  $\text{LaCrO}_3$  type. By modifying the stoichiometry with other elements it is possible to adapt this interconnect material with respect to thermal expansion and behavior in the presence of reaction gases [48,53]. However, the material costs of perovskites are rather high and their application as ceramic interconnect is only meaningful as long as the stack design requires only small amounts of the material as in the case of the tubular system of MHI (Table 4). Furthermore, lanthanum chromites are often plasma-sprayed (Table 4) although this technique is expensive. In this case, however, one has to consider the frequently observed low sinterability of the interconnect material, which prevents gastightness and low-cost production by sintering since it requires sintering temperatures between 1450 and 1600 °C.

Whereas practically all activities are related to  $\text{LaCrO}_3$  modifications for the ceramic interconnects, clearly more material systems are under development for the metallic interconnects. In general, advantages for metallic arrangements are considered to be high electrical conductivity, good processability and the lower costs to be expected, whereas especially long-term resistance, corrosion behavior, chromium evaporation and high expansion coefficients are disadvantageous. The success of metallic interconnects for use in the SOFC system will decisively depend on solving of these problems.

The long-term stability of the metallic interconnect is essentially governed by its corrosion characteristics. The materials used for interconnects are chromia forming alloys which ensure sufficiently high conductivity for thin oxide scales.

With respect to thermal expansion, electrical conductivity and corrosion behavior, the Cr 5Fe 1Y<sub>2</sub>O<sub>3</sub> ODS alloy developed by Plansee in cooperation with Siemens shows excellent behavior at temperatures up to 950 °C [54]. A disadvantage of the alloy produced by powder metallurgy is the currently high price which could be drastically reduced by suitable production techniques. A relevant approach is near-net-shape processing producing the interconnects practically without a finishing operation [55].

From the aspect of costs, ferritic chromium steels are attractive candidates for metallic bipolar plates. On the one hand, they form chromium oxides, have a lower thermal expansion compared to austenitic alloys and can be mechanically easily deformed and machined. On the other hand, they have a number of properties limiting their application such as lower high-temperature strength, insufficient corrosion protection at high temperatures and brittle phase formation. R&D work on ferritic steels therefore concentrates on application temperatures <800 °C. The application range of interest for this material class coincides with the development goals for planar anode-supported fuel cells. For this reason, such materials are being used or developed by all companies and research institutions working on this concept (e.g. Sulzer, CFCL, Plansee, Sanyo, FZJ) [56–58]. In general, it can be stated that the long-term corrosion behavior of commercially available materials is not yet sufficient. Recent developments [59] have attempted to achieve acceptable long-term behavior by selected doping with reactive elements (Ti, Y, La) and spinel formers.

In operating high-temperature fuel cells with metallic interconnects, a time-dependent degradation is observed, which is attributable to poisoning of the active centers of the cathode by chromium evaporating from the interconnect. This familiar process, already described in detail [60–62], is caused by highly volatile chromium species which form when chromium oxide is in contact with oxidizing gas atmospheres. CrO<sub>2</sub>(OH)<sub>2</sub> and CrO<sub>3</sub> are particularly critical here [60] and can react with the perovskite of the cathode to more stable but less catalytically active Cr–Mn spinels [62].

Attempts are being made to reduce the damaging effect of chromium vapors on the cathode side by suitable protective layers of LaCrO<sub>3</sub> or by neutraliz-

ing the chromium atoms in the applied layers by gettering. This development is most advanced for the Cr5Fe1Y<sub>2</sub>O<sub>3</sub> chromium-base interconnect, on which La–Sr–Cr oxides have yielded the best results [63]. Vacuum plasma spraying and physical vapor deposition have proved to be effective application processes [64,65]. The best result so far has been obtained with a La<sub>0.9</sub>Sr<sub>0.1</sub>CrO<sub>3</sub> coating for operation temperatures of about 950 °C. At a temperature of 850 °C, the chromium vaporization rate was only half as great [66]. Future further developments and tests must above all demonstrate the long-term stability and thermal cycling resistance of such coatings.

Except for the Cr-base material, the metallic interconnects are manufactured using the casting-rolling-(forging) route with mechanical machining of the semi-finished products. The solutions have not yet reached the cost goal. Intensive work is being performed to find cheaper solutions by simplifying and adapting the design and using other modern manufacturing techniques (e.g. punching, laser cutting, brazing, etc.).

## 5. Summary

The overview of this report shows the wide diversification in SOFC technology. The manufacturing and processing of materials and components has reached a converging stage where technologies with a known potential for mass production and cost reduction are preferred. For systems close to market launch, electrode and electrolyte materials are favored which have been well known and investigated for several decades and which have demonstrated the best long-term stability and reliability. Other materials and fabrication processes needed for stacking and construction have to be tested in more detail to ascertain whether they can fulfil all the requirements for such a complicated joined system.

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