

# Materials and concepts for solid oxide fuel cells (SOFCs) in stationary and mobile applications

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## Abstract

Solid oxide fuel cells (SOFCs) are a forward-looking technology for highly efficient, environmentally-friendly power generation. The electrochemically active single cell is a multilayer structure consisting of ceramic and metallic materials with different electrical transport properties. Materials for electrodes should exhibit electronic and ionic conductivity in combination with porosity and catalytic activity, whereas the electrolyte has to be a purely ionically conducting and gas-tight membrane. All components have to show well-adjusted thermal expansion behavior, chemical compatibility of material interfaces and chemical stability in the prevailing temperature and gas atmosphere. The performance and long-term stability of the single cell is significantly increased by the use of suitable materials, a proper design of the cell and an optimized microstructure at the electrode/electrolyte interfaces. The application of appropriate technologies for the production of single cells with optimized microstructure becomes even more important for highly efficient SOFCs operating in the medium and low temperature range.

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## 1. Introduction

Starting from the different available fuels like natural gas or heating oil for stationary applications as well as gasoline or diesel fuel for propulsion, solid oxide fuel cells (SOFCs) offer significant advantages for a high efficient electrochemical energy conversion. Due to their high operating temperatures in-between 600 °C and 1000 °C a wide variety of fuels can be processed [1–3]. A substantial progress in materials, fabrication and system-technology enabled field tests of different SOFC systems within the last years. Up to now only small stationary SOFC units in the power range of 1 kW to less than 300 kW have been realized. But even the small systems are capable of electrical net efficiencies up to 50% [4–6] and overall efficiencies of about 70–90%. Large pressurized SOFC/gas-turbine systems in the MW-range are expected to achieve electrical net efficiencies up to 70% [7]. The stationary application of SOFC in small combined heat and power generation systems for one-family houses is already on the way to the market, the setup of large scale production capacities within the next years is expected to result in a significant cost reduction [8,6].

Due to the lack of a hydrogen infrastructure and the technical effort required for on board hydrogen production, which is necessary for PEMFC powered electric vehicles, mobile applications of SOFC are attracting more and more interest. The development of SOFC-based auxiliary power units (APU) for cars, trucks (BMW/Delphi) [9,10] and military applications [11] as well as the development of low cost, high power density SOFC core modules which can be used in stationary as well as in a mobile systems (solid state energy conversion alliance: SECA) broadens the areas of SOFC application and enhances the demands on SOFC materials and components (Fig. 1).

The high operating temperature of SOFCs is advantageous for the processing of common fuels and for combined SOFC/gas-turbine power plants. On the other hand, significant disadvantages concerning the long-term stability and costs of materials and components as well as the demand for a short startup time in mobile applications arise.

The operating temperature of a SOFC is restricted by thermally activated transport processes and electrochemical reactions such as the oxide ion conductivity of the solid electrolyte and different reaction steps in the electrodes, respectively, at the electrode/electrolyte interfaces.

Decreasing the operating temperature generally results in decreased power density and/or efficiency. On the other hand, the long-term stability of the system can be improved

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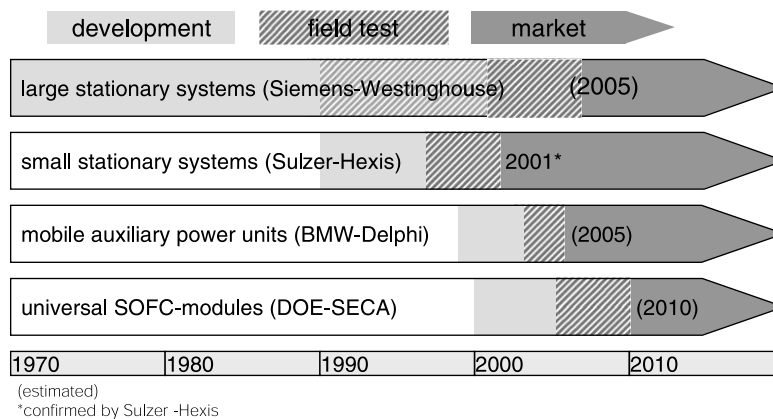


Fig. 1. Development status and targets for different SOFC applications.

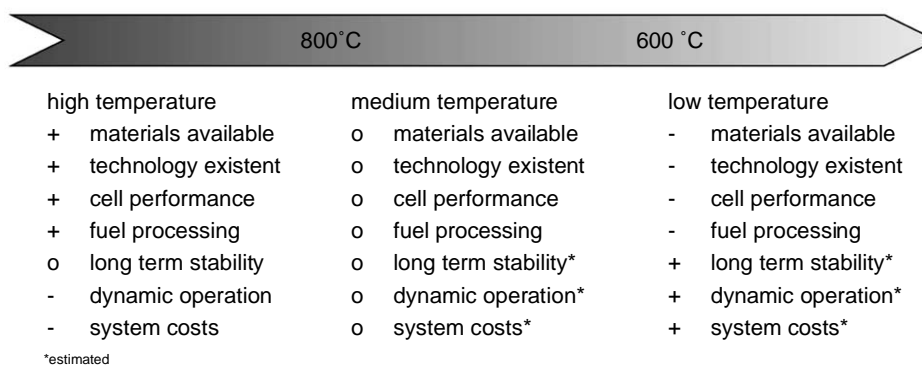


Fig. 2. Advantages (+) and disadvantages (-) of SOFCs for different temperature ranges: (O) represents states between (+) and (-).

and the system costs reduced by using less costly metal alloys for interconnects and external components. Therefore the improvement of cells and stacks for an intermediate temperature SOFC operating economically in-between 600 and 800 °C is required whereas for temperatures below 600 °C new materials, cell concepts, production technologies and suitable stack designer have to be developed (Fig. 2).

Stationary SOFCs operating in the high temperature range meet the requirements concerning cell performance. Suitable materials and production technologies are available. The system costs have to be decreased decisively and the long-term stability of cells and stacks has to be improved. Dynamic operation, i.e. fast startup, load and thermal cycling as well as redox-cycles due to gas supply failures are still unsolved problems. For intermediate operating temperatures (600–800 °C) the development targets concerning power density and efficiency of single cells are almost fulfilled [12–14].

## 2. Electrolyte-supported cells for high operating temperatures

A SOFC is a multilayer structure consisting of ceramic and metallic materials. There are different types of SOFC

concepts that differ in the single cell design and arrangement, interconnector materials and gas flow. SOFC materials have to fulfill different requirements. The electrolyte has to be a gas-tight and purely ionic conducting membrane whereas the electrodes have to enable the transport of electrons as well as of gaseous reactants and reaction products and therefore have to be porous. The electrode materials should exhibit a high catalytic activity for the desired chemical and electrochemical reactions. The microstructure of the electrodes should display a large number of active reaction sites. All materials and components of the stack have to show chemical stability in the prevailing atmospheres whereupon changes in the gas composition due to fuel and oxygen utilization, fuel supply failures and leakage in the stack have to be taken into account. The compatibility in-between the different materials, i.e. a well-adjusted thermal expansion behavior, chemical compatibility and good adhesion at the interfaces has to be fulfilled.

State of the art materials currently used in most SOFC systems are yttria stabilized zirconia (YSZ) as the electrolyte which can be used either as TZP (3YSZ: ZrO<sub>2</sub> doped with ~3 mol% Y<sub>2</sub>O<sub>3</sub>) or CSZ (8YSZ: ZrO<sub>2</sub> doped with ~8 mol% Y<sub>2</sub>O<sub>3</sub>). Although the ionic conductivity of TZP is significantly lower, this material is advantageous because of its outstanding mechanical stability. Strontium doped lanthanum

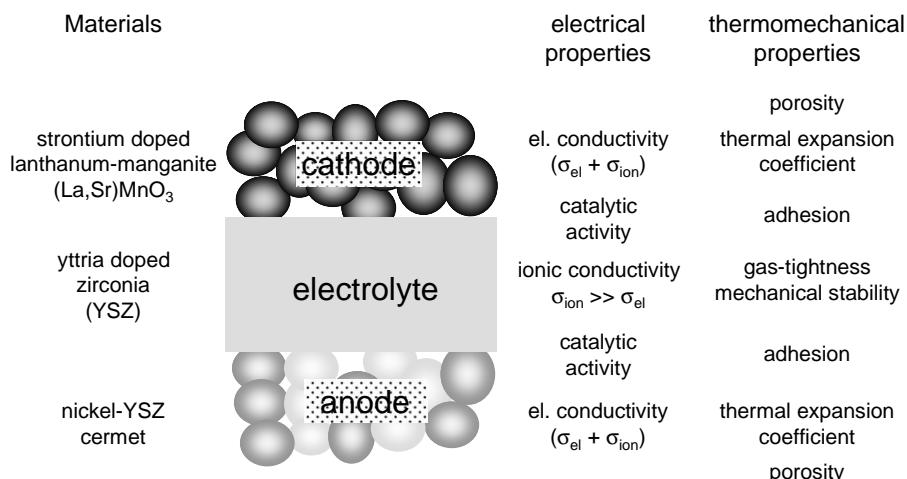


Fig. 3. State of the art materials and required electrical and (thermo) mechanical properties of SOFC-single cells.

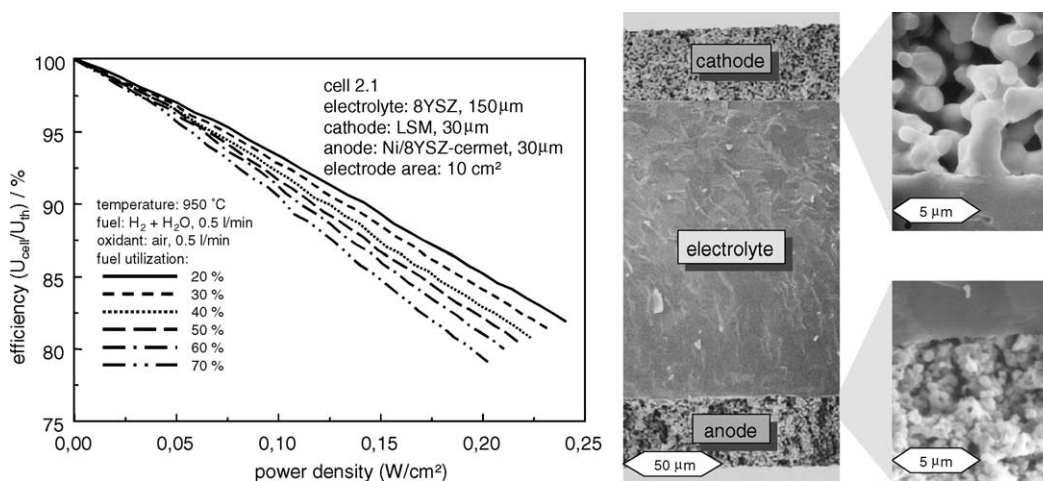


Fig. 4. Nernst efficiency vs. power density at different fuel utilization and SEM images of an electrolyte-supported single cell with monolayer electrodes.

manganite (LSM) is used as the cathode and nickel/YSZ cermets as the anode (Fig. 3).

The electrical efficiency (cell voltage/open circuit voltage, fuel efficiency not included) as a function of power density of a planar single cell with monolayer electrodes is given in Fig. 4. This type of cell just fulfills the targets concerning power density (Table 1).

An investigation of the loss mechanisms in this type of single cell by in situ impedance spectroscopy as well as the use of reference electrodes revealed that the cathode governs the main part of the losses. In the case of pure electronic conducting electrode materials like metals or some perovskite type oxides (LSM), the electrochemical reactions are almost restricted to the triple phase boundaries (tpb) [15–17]. The transport of oxide ions within the electrode material is advantageous concerning the number of possible reaction pathways. Therefore electrodes should be either a composite consisting of an electronic and an ionic conducting phase or a mixed conducting metal oxide to enlarge the active area into the electrode volume (Fig. 5).

To increase the cell performance of an electrolyte-supported single cell the polarization losses of the electrodes, mainly of the cathode, have to be decreased. On the one hand, the electrochemical properties of the cathode can be influenced by choosing an appropriate composi-

Table 1  
Target areas for mobile and stationary SOFC systems

Application	Stationary	Mobile (traction)
Power density (W/cm <sup>2</sup> )	>0.25	>1 <sup>a</sup>
Operation temperature (°C)	700–1000	500–700
Lifetime (operation) (h)	>40000	>2000
Degradation rate (µV/h)	<1	<10 <sup>a</sup>
Fuel utilization (%)	>80	>80 <sup>a</sup>
Thermal cycles	>100 <sup>a</sup>	>5000 <sup>a</sup>
Heating rates (K/min)	>1	>100 <sup>a</sup>
Fuel	Natural gas, fuel oil	Gasoline, diesel
Oxidant	Air	Air
System costs (\$/kW)	<500	<100

<sup>a</sup> Estimated.

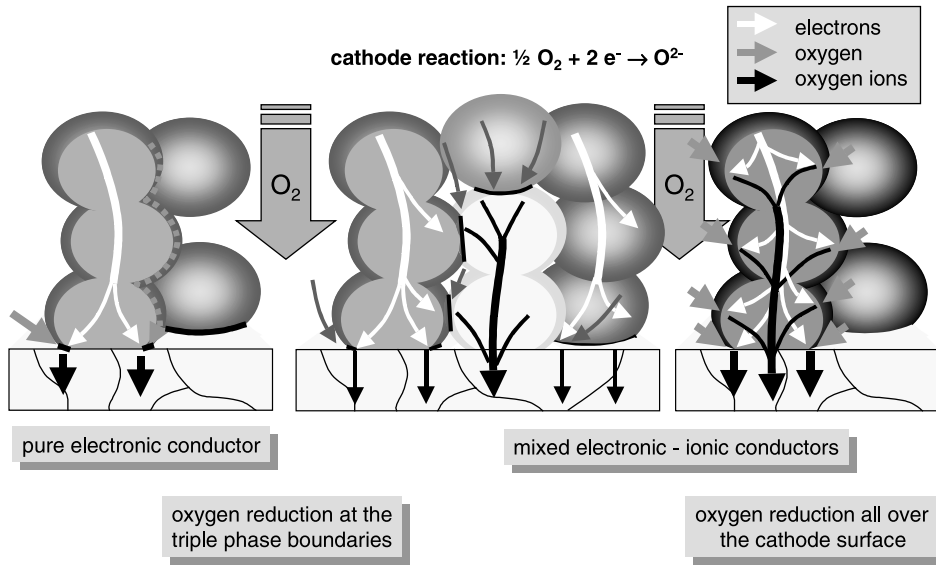


Fig. 5. Oxygen reduction at a pure electronic, composite and mixed conducting cathode.

tion. Different perovskite type oxides  $ABO_3$  exhibiting lanthanum and strontium or calcium as a dopant on the A-site and transition metals like Cr, Mn, Fe, Co and/or Ni on the B-site have been investigated with respect to their suitability as a cathode material [18,19]. For example, in the  $La_{0.8}Sr_{0.2}Mn_{1-x}Co_xO_{3-d}$  solid solution (LSMC), the electrical and oxygen ion conductivity can be increased significantly by substituting a part of the manganese with cobalt [20,21] (Fig. 6). Therefore the use of this type of cobalt-containing cathode materials should result in a decreased cathode polarization resistance. On the other hand, a high amount of cobalt results in an increased thermal expansion coefficient (TEC) resulting in a delamination at the cathode/electrolyte interface or a cracking of the electrolyte.

A well-adapted thermal expansion behavior can be reached by using either an alternative mixed conducting cathode material (Fig. 7) or a composite consisting of

a mixed conducting cathode material and an appropriate amount of electrolyte material.

A severe problem of LSMC and similar cathodes is the insufficient chemical compatibility between these cobalt-containing cathode materials and YSZ [22]. Sintering of the cathode-layer onto the electrolyte substrate at temperatures  $>1100^\circ\text{C}$  leads to the formation of secondary phases like lanthanum- and strontium-zirconate (Fig. 8). These insulating interlayers lead to a significant increase of the cathodic polarization resistance [23].

Other possibilities to decrease the cathodic polarization losses are the improvement of the microstructure at the cathode/electrolyte interface as well as the enlargement of the effective cathode/electrolyte interface area. A significant increase of the cell performance is possible using an improved interface as shown in Fig. 9 [24]. The use of metal organic deposition (MOD) technology for applying nanoporous thin

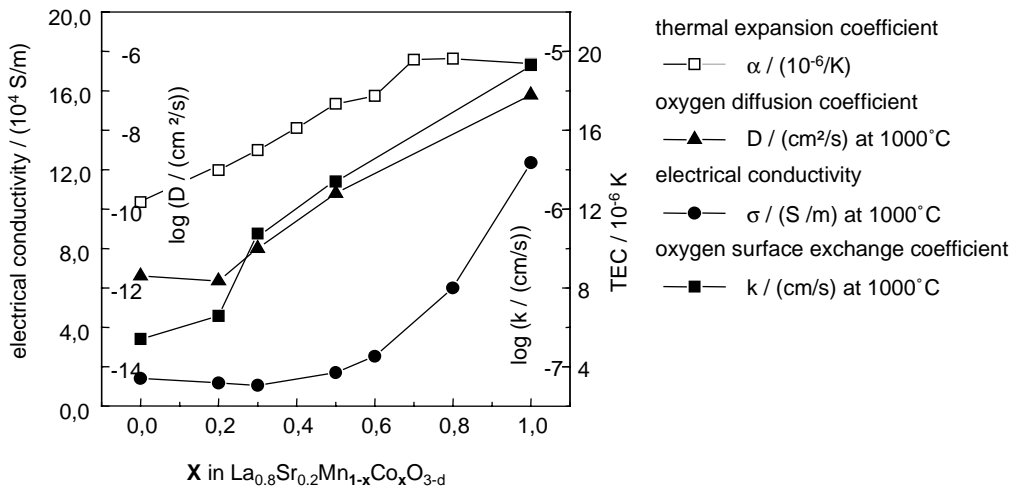


Fig. 6. Electrical and thermomechanical properties of the  $La_{0.8}Sr_{0.2}Mn_{1-x}Co_xO_{3-d}$  solid solution (Siemens/Imperial College).

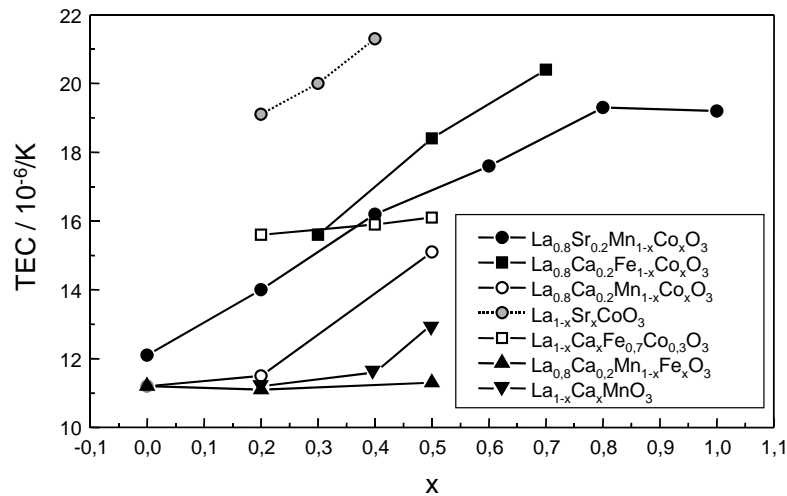


Fig. 7. Thermal expansion coefficient (TEC) of various cathode materials.

film cathodes offers significant advantages. Due to the low processing temperatures and times ( $\ll 1000^\circ\text{C}$ , rapid thermal annealing process), the formation of secondary phases in-between cathode and electrolyte is prevented. The application of mixed conducting cathode materials like strontium doped lanthanum cobaltate (LSC) becomes possible [25].

The performance of electrolyte-supported single cells made of a  $150\ \mu\text{m}$  thick 8YSZ-substrate and a Ni/YSZ-anode with different types of cathodes is shown in Fig. 10. In spite of the advantageous material properties of LSC, which exhibits a significantly higher electronic and ionic conductivity than LSM (Fig. 6), the cell with a monolayer LSC-cathode shows lowest performance. Applying a structured electrolyte surface and a LSM MOD thin film cathode a decisive enhancement in performance was achieved. In case of a LSC MOD thin film cathode, an additional increase in performance is possible.

Besides an increase in performance, single cells with this type of optimized cathode/electrolyte interface show an enhanced long-term and thermocycling stability. Whereas a monolayer cathode shows extensive delamination after a few thermocycles, the three dimensional penetration structure between electrolyte and cathode inhibits any significant delamination. The thermocycling stability, i.e. the increase in cell resistance due to thermal cycling is shown in Fig. 11.

Nevertheless degradation is a serious problem at high operating temperatures. Depending on the operating conditions, a gradient in oxygen chemical potential at the cathode/electrolyte interface (LSM-cathode/YSZ electrolyte) might induce a demixing of the materials resulting in microstructural changes and a minor adhesion. Even at moderate operating conditions ( $850^\circ\text{C}$ ,  $200\ \text{mA}/\text{cm}^2$  for 4500 h, Fig. 12) a depletion of yttrium and an interdiffusion of manganese as well as the formation of

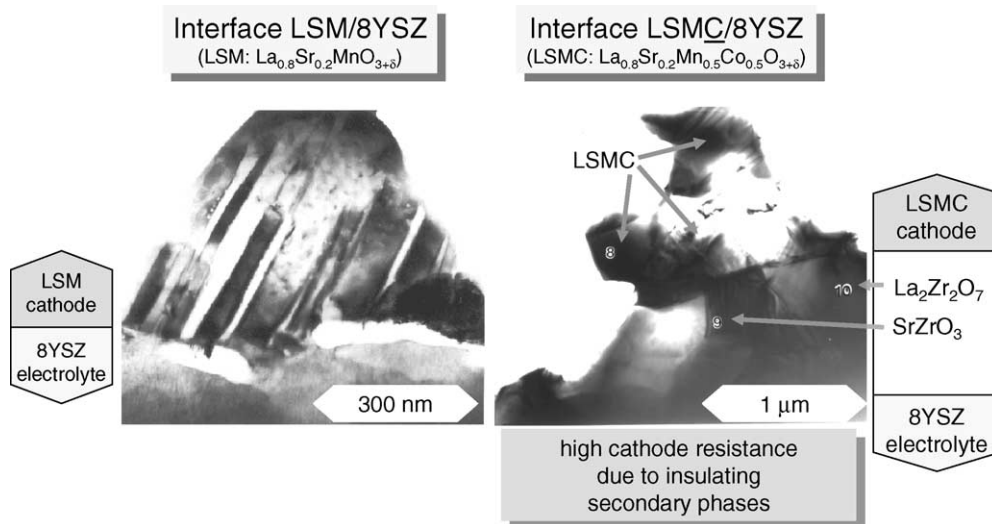


Fig. 8. TEM images of cathode/electrolyte interfaces. The interface between LSM and 8YSZ shows no secondary phases at all whereas in-between the cobalt-containing LSCM cathode and the 8YSZ electrolyte the formation of secondary phases (lanthanum- and strontium-zirconate) takes place.

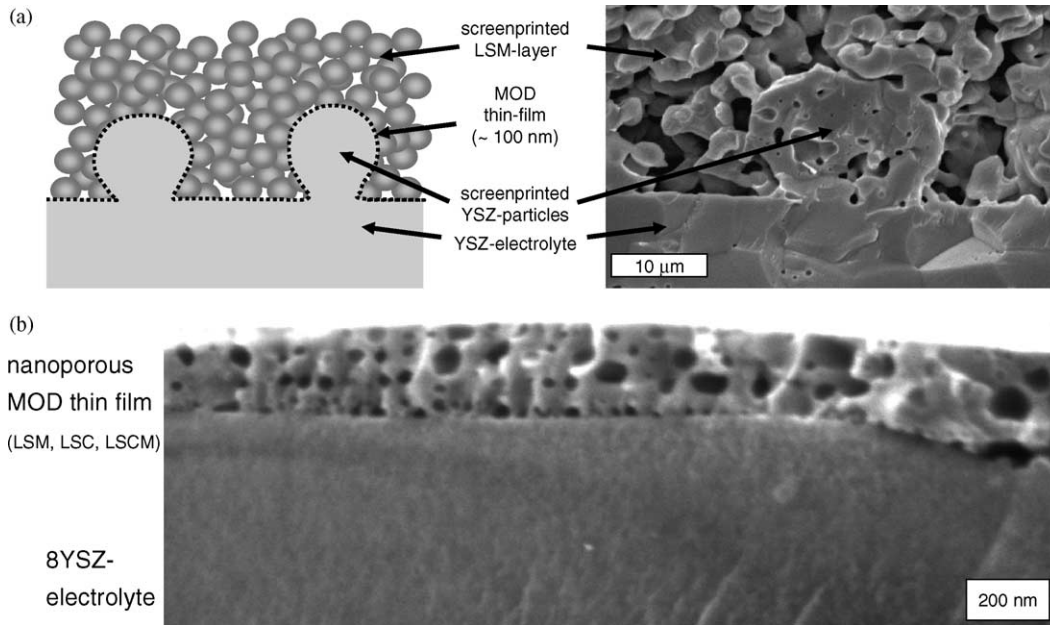


Fig. 9. Improved cathode/electrolyte interface: the effective electrolyte surface area is enlarged by (a) structuring the substrate surface with screenprinted 8YSZ particles; (b) coating the whole electrolyte surface with a nanoporous electrochemical active MOD, LSM, LSCM or LSC thin film. The screenprinted LSM layer on top performs only as a current collector and gas distribution layer.

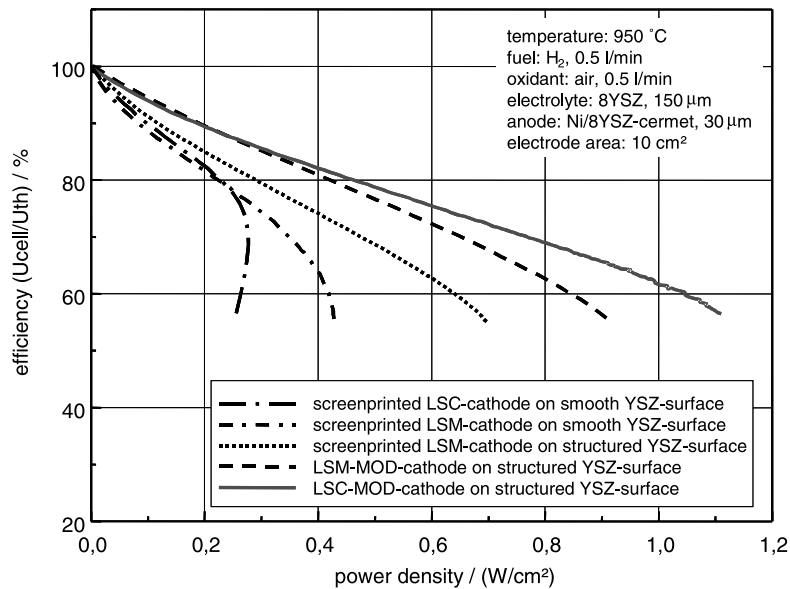


Fig. 10. Efficiency vs. power density of electrolyte-supported single cells with different cathodes.

micropores at the LSM/YSZ interface was observed [26].

### 3. Cells for medium and low operating temperature

Due to significant problems concerning long-term stability of cells and other stack components for SOFC systems operating at high temperatures, an operating temperature in-between 600 and 800 °C exhibits many advantages. The temperature dependence of cell volt-

age and losses of an electrolyte-supported single cell are shown in Fig. 13. This type of standard cell, consisting of an 8YSZ electrolyte with screenprinted LSM monolayer cathode and Ni/YSZ cermet anode, exhibits insufficient performance operating at temperatures <850 °C.

Intermediate operating temperatures result in a drastic increase in the internal cell resistance. For reduced operating temperatures the polarization losses, especially at the cathode, and the ohmic losses in the electrolyte have to be lowered substantially.

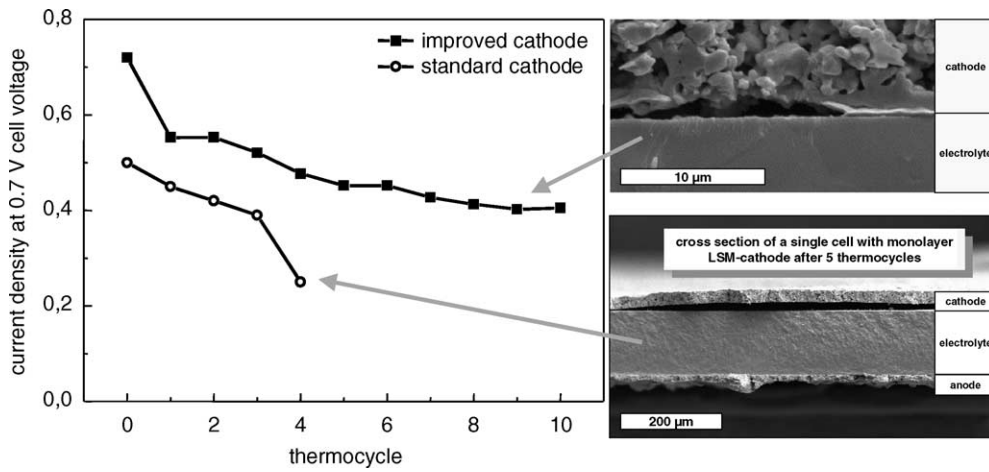


Fig. 11. Decrease in cell performance due to thermal cycling of single cells during operation. Whereas the cell with a standard monolayer cathode fails after five thermocycles, the improved cathode shows an enhanced stability (the decreased cell performance includes degradation of electrolyte and anode).

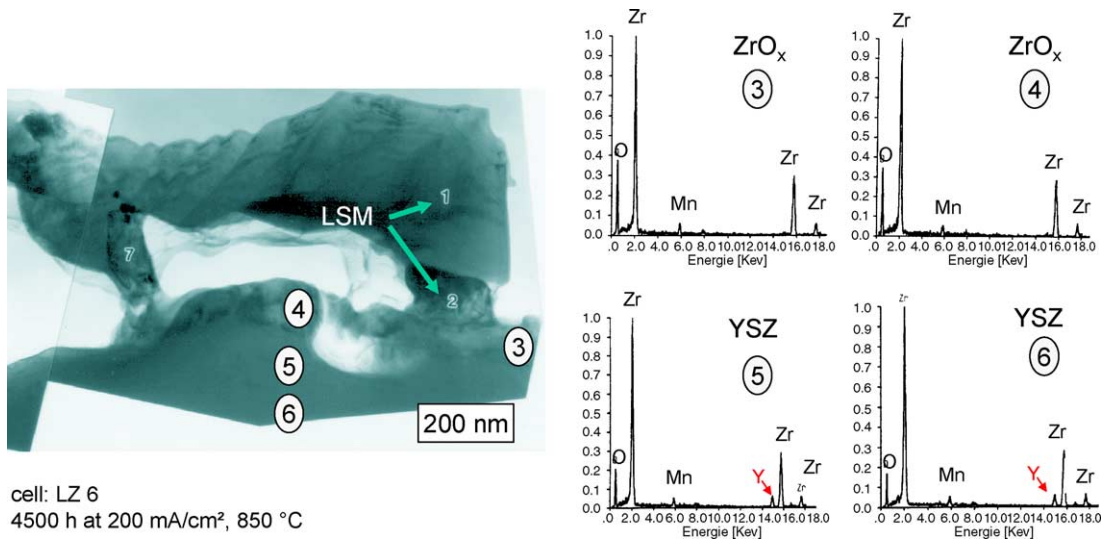


Fig. 12. TEM and EDX analysis of the LSM/8YSZ interface after 4500 h of operation at 850 °C and 200 mA/cm<sup>2</sup> revealed Mn-diffusion and Y-depletion at the surface of the 8YSZ electrolyte.

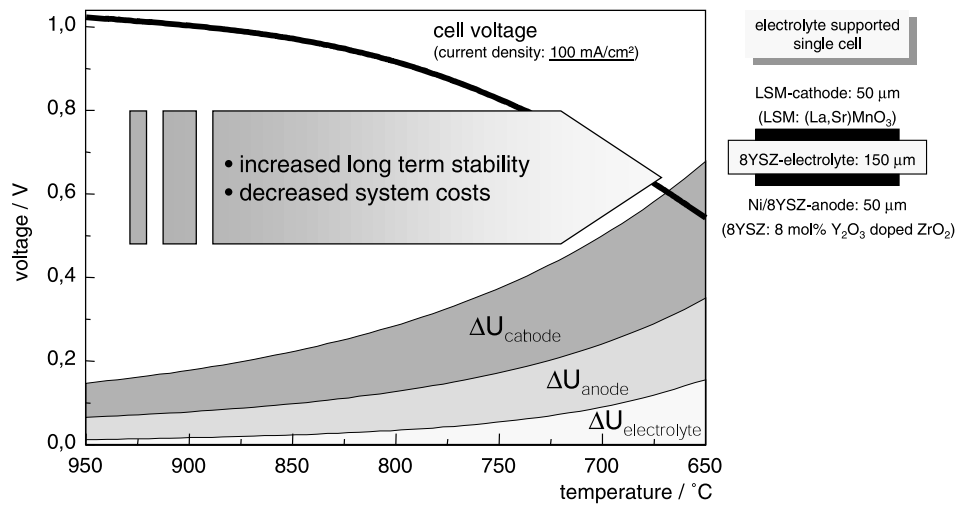


Fig. 13. Cell voltage of an electrolyte-supported single cell (150 μm 8YSZ electrolyte substrate, screenprinted monolayer electrodes) at a constant current density of 100 mA/cm<sup>2</sup> as a function of operating temperature (a sufficient power density is achieved at an average current density of about 300 mA/cm<sup>2</sup>).

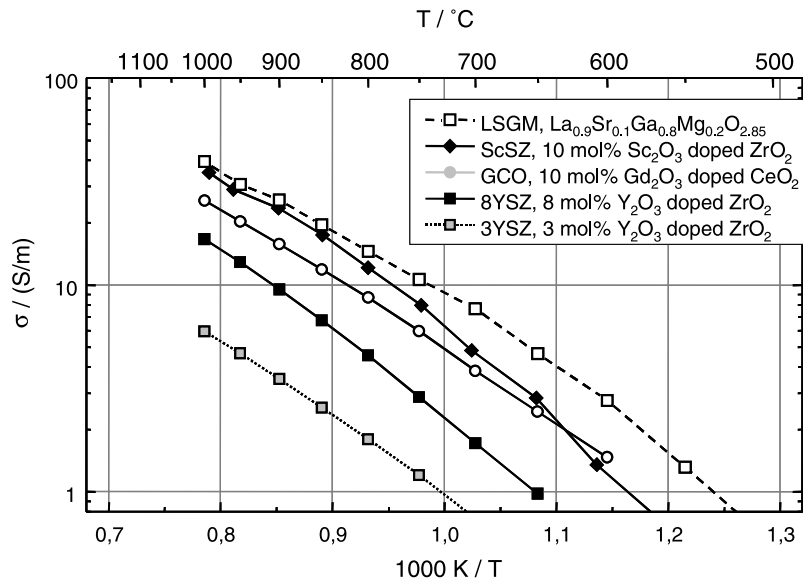


Fig. 14. Oxide ion conductivity of different electrolyte materials [34,35].

The electrolyte resistance can be reduced using an alternative electrolyte material with higher oxide ion conductivity than YSZ or/and an electrode-supported thin film electrolyte. Beneath a high oxide ion conductivity additional requirements for medium and low temperature solid electrolytes have to be fulfilled. The material must exhibit pure oxide ion conductivity, i.e. no electronic conductivity and structural as well as mechanical stability in a wide temperature and oxygen partial pressure range. In Fig. 14

the conductivity of various candidate materials for solid electrolytes that have been studied by many research groups intensively is given as a function of temperature.

Ce-based oxide ion conductors like Gd doped CeO<sub>2</sub> (GCO) offer a significantly higher ionic conductivity but they become mixed conductors in the anode environment resulting in an internal short circuit of the cell. Therefore a decreased open circuit voltage (OCV) and additional fuel utilization even under OCV conditions takes place resulting

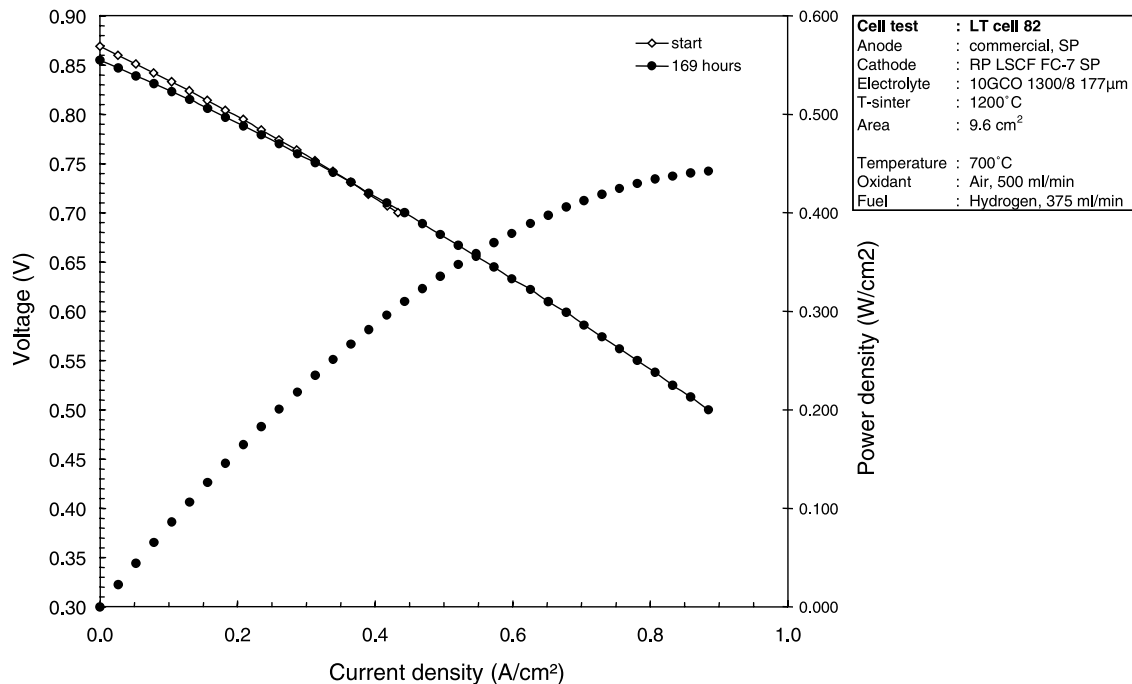


Fig. 15. C/V characteristics of a GCO electrolyte-based single cell at 700 °C (ECN).



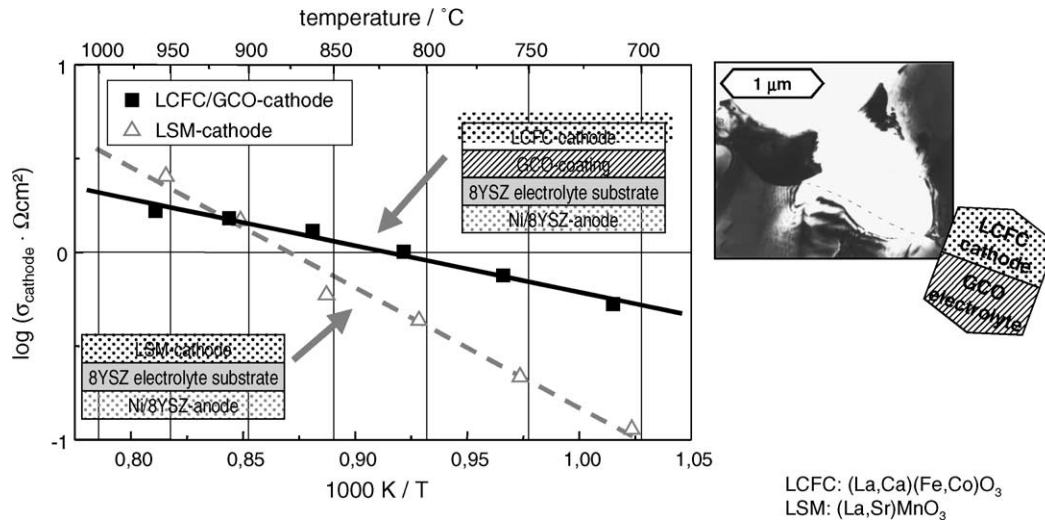


Fig. 16. Temperature dependence of the cathode resistance of single cells with electronic conducting cathode (screenprinted LSM layer on 8YSZ-substrate) and mixed conducting cathode (screenprinted LCFC layer + GCO interlayer on 8YSZ-substrate).

in a decreased system efficiency [27]. Nevertheless the application of GCO electrolytes at operating temperatures <700 °C is possible. Due to the excellent chemical compatibility with mixed conducting cathode materials, the power density and efficiency of a GCO electrolyte-based cell might be sufficient (Fig. 15).

Another possibility is the use of double- or multi-layer electrolytes. To take advantage of the compatibility of Ce-based electrolytes with mixed conducting cathode materials and the mixed conducting properties of doped CeO<sub>2</sub> in an anode environment, interlayers and composite or cermet electrodes containing doped CeO<sub>2</sub> can be used (Fig. 16) [28,29]. Lanthanum Gallate (LSGM (La, Sr)(Ga, Mg)O<sub>3</sub>)-based electrolytes exhibit the highest oxide ion conductivity of possible electrolyte materials for SOFC so far [30,31]. LSGM also shows an excellent chemical compatibility with mixed conducting cathode materials. The long-term stability due to Ga-evaporation in a reducing atmosphere and the compatibility with Ni seem to be severe problems at high operating temperatures [32]. The use of LSGM-electrolyte substrates contains problems due to the low mechanical stability of LSGM and the high costs of gallium. LSGM might be an interest-

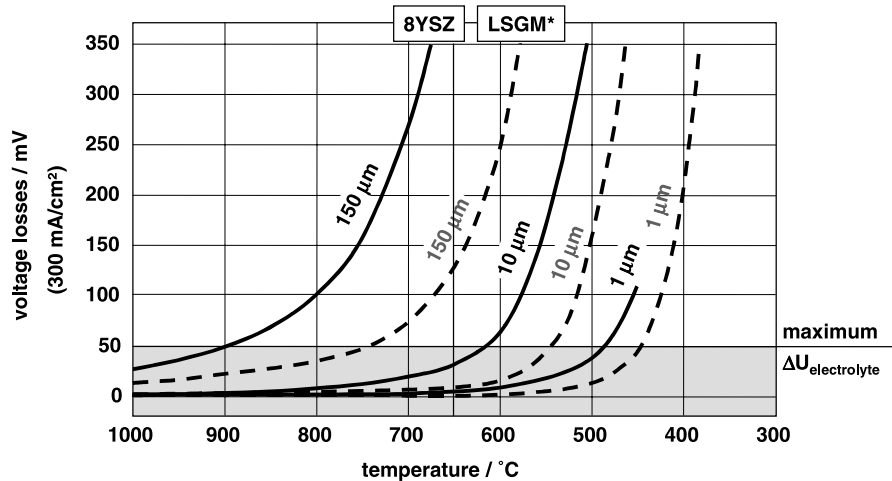
ing electrolyte material for low operating temperatures (<600 °C).

Table 2 gives an overview of the advantages and disadvantages of the different electrolyte materials. With respect to all secondary requirements for an electrolyte material, it is still questionable if zirconia-based electrolytes can be replaced. A way out might be doping zirconia with scandium instead of yttrium. Scandium doped ZrO<sub>2</sub> (ScSZ) exhibits a significantly higher ionic conductivity than YSZ. Sc-doped ZrO<sub>2</sub> electrolytes are the most promising alternatives for intermediate and even low operating temperatures [33]. But up to now the price and availability of scandium is uncertain and there is no high quality ScSZ powder available yet.

Another possibility to increase the cell performance at decreased operating temperatures is a reduction of the electrolyte thickness. For electrolyte-supported cells the minimum thickness is in-between 150 and 250 μm for CSZ and ceria-based materials. Due to the outstanding mechanical stability of TZP, a minimum thickness of only 50 μm for TZP seems to be possible. In the case of a supported thin film electrolyte, a thickness of a few microns is possible even for large cell areas. The voltage losses in the electrolyte at an acceptable current density of 300 mA/cm<sup>2</sup> as a function of

Table 2  
 Advantages and disadvantages of possible electrolyte candidates for SOFC

YSZ	GCO	LSGM	ScSZ
Excellent stability in oxidizing and reducing environment excellent mechanical stability (3YSZ) >40,000h of fuel cell operation possible high quality raw materials available	Good compatibility with cathode materials mixed electronic-ionic conductor at low pO <sub>2</sub> (application in anode cermets)	Good compatibility with cathode materials	Excellent stability in oxidizing and reducing environment better long-term stability than 8YSZ?
Low ionic conductivity (especially 3YSZ) incompatible with some cathode materials	Electronic conduction at low pO <sub>2</sub> → low OCV mechanical stability availability and price of Gd	Phase stability, Ga-evaporation at low pO <sub>2</sub> incompatible with NiO mechanical stability availability and price of Ga	Availability and price of Sc



\* conductivity values: T. Ishihara et al., Proc. 5<sup>th</sup> Intl. Symp. SOFC, The Electrochemical Society, 301-310, (1997).

Fig. 17. Electrolyte losses as a function of operating temperature and electrolyte thickness for 8YSZ and LSGM (current constriction ignored).

temperature and electrolyte thickness are shown in Fig. 17 for 8YSZ and LSGM.

Accepting electrolyte losses <50 mV, the lowest operating temperature of electrolyte-supported single cells (LSGM or ScSZ electrolyte substrate, 150 μm thickness) is about 750 °C, whereas a supported thin film electrolyte theoretically exhibits a minimum operating temperature of less than 500 °C. Including the polarization losses at the electrodes a sufficient cell performance can be achieved at about 650–750 °C using the state of the art materials (Fig. 18). The different electrolyte and electrode-supported planar single cell concepts are shown in Fig. 19. The most common is the anode supported one (Allied Signal, FZ-Jülich and many others); the most advanced tubular Siemens–Westinghouse concept is based on a cathode-supported electrolyte. An advantage of all cathode-supported single cells is a three-dimensional penetration structure in-between cathode and electrolyte that is achieved during electrolyte deposition. With state of the art materials in combination with an ScSZ thin film electrolyte, polarization losses can be reduced significantly at intermediate operating temperatures.

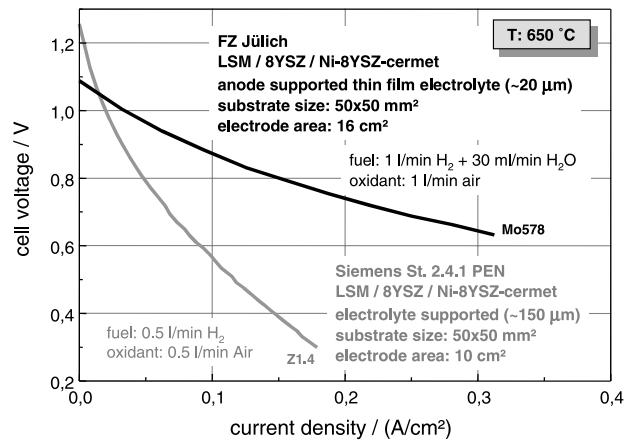


Fig. 18. Current/voltage characteristics of electrolyte and anode supported single cells consisting of state of the art materials (LSM/8YSZ/Ni-8YSZ) at 650 °C.

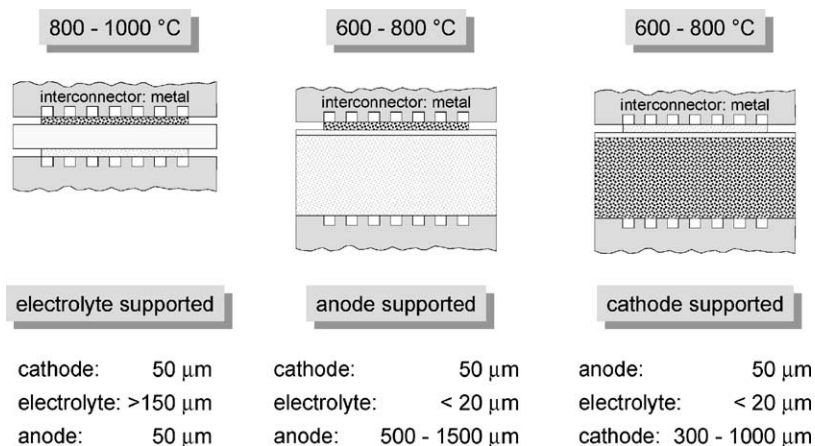


Fig. 19. Planar single cell concepts.

#### 4. Conclusions

The optimum choice of materials, technology and single cell design for SOFC depends significantly on the stack design and the system requirements. Even at high operating temperatures, the well-proven tubular Siemens–Westinghouse concept has to be followed by an alternative SOFC concept due to the demands of cost reduction and power density increase. For intermediate and low operating temperatures in small-scale stationary and mobile application the following issues can be stated:

- With respect to the electrical performance, electrolyte-supported single cells with ScSZ electrolyte and custom-tailored electrode/electrolyte interfaces using well established anode cermets and mixed conducting cathode materials are promising to temperatures as low as 750 °C.
- Below 750 °C thin film electrolytes (probably still zirconia-based) have to be applied.
- The interfaces between both electrodes and the thin film electrolyte have to be optimized for low polarization losses and high long-term stability independent of the chemical composition of the electrode materials.
- Long-term degradation effects due to interdiffusion and demixing will be less important but have to be considered as well.
- With respect to thermomechanical failures, all kinds of thin films, electrode/electrolyte interfaces and support materials have to be well adapted in their thermal expansion behavior and mechanical properties.
- The dynamic properties of cells and stacks, i.e. electrical loading, fast startup/cool down procedures and the redox stability of the anode has to be improved.
- The requirements for single cells and stacks are determined by the application. Stationary and mobile SOFC systems have to fulfill different properties; materials, cells and stacks have to be custom-tailored.

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