

Available online at www.sciencedirect.com



Journal of Power Sources 138 (2004) 111-119



www.elsevier.com/locate/jpowsour

# Deformation behavior and leakage tests of alternate sealing materials for SOFC stacks

Martin Bram<sup>\*</sup>, Stephan Reckers, Pere Drinovac, Josef Mönch, Rolf W. Steinbrech, Hans Peter Buchkremer, Detlev Stöver

Forschungszentrum Jülich, Institute for Materials and Processes in Energy Systems IWV, Jülich, Germany

Received 4 March 2004; accepted 21 June 2004 Available online 14 August 2004

## Abstract

Compression seals based on metal/mica composites were investigated under solid oxide fuel cell (SOFC) operating conditions. Since a compressive force has to be permanently applied in this sealing concept, the mechanical properties of the sealing materials and the SOFC components also became important for maintaining gas tightness. Leakage tests with a sandwich arrangement of mica paper and embossed metallic profiles show leak rates of  $<1 \times 10^{-4}$  mbar l/mm s under an applied normalized compressive load of 2.7 N/mm (0.7 MPa). Also under thermal cycles between ambient temperature and 800 °C this leak rate does not change due to the elastic response of the seal. Compression tests with the seals in contact with SOFC membranes reveal no damage to the membranes. First results of leakage tests with SOFC stacks are presented, which may serve as a reference for future leakage tests of alternative sealing systems. © 2004 Elsevier B.V. All rights reserved.

Keywords: Solid oxide fuel cells; Compressive metallic seals; Mica; Mechanical properties; Leakage rate; Stack testing

# 1. Introduction

Stacks of planar solid oxide fuel cells (SOFC) are believed to offer the potential for higher cost efficiency and power density per unit volume when compared to, for example tubular designs. However, the high-temperature sealing concept in planar design is one of the crucial challenges of this power conversion technology. The occurrence of leakage during long-term operation of a stack may contribute to the degradation of the electrical stack performance. In particular, local sealing failure must be strictly prevented because the severe exothermal reaction between fuel and oxidant significantly increases the temperature. As an extreme, the socalled hot spots can cause a catastrophic loss of electrical performance during stack operation.

Seals in SOFC stacks have to fulfil a variety of requirements. Primarily, long-term stable separation of oxidant and fuel gases, even under thermal cycling operation, has to be achieved. Thermochemical and thermomechanical compatibility with the adjacent SOFC components should be realized. The sealing materials should not affect or deteriorate the brittle electrolyte (8 mol.% yttria-stabilized zirconia 8YSZ) and the interconnect steel (chromium alloyed ferritic steel). Furthermore, electric insulation is required to avoid a short circuit when joining the interconnects. Depending on the design, geometrical limitation of the sealing components, e.g. corners and junctions, have to be taken into account. Adequate sealing should also sufficiently compensate the mechanical and thermal mismatch of the stack components. In the case of metallic seals, the creep behavior and the elastic recovery are important.

Currently glass-ceramics are the preferred materials in many rigid sealing concepts of SOFC stacks [1–4]. They combine moderate sealing loads with few restrictions in stack design and provide sufficient gas tightness for mid-term stack operation (several 1000 h). The thermal expansion coefficient (TEC) of glass-ceramics can be adapted to other stack com-

<sup>\*</sup> Corresponding author. Tel.: +49 2461 616858; fax: +49 2461 612455. *E-mail address:* m.bram@fz-juelich.de (M. Bram).

<sup>0378-7753/\$ –</sup> see front matter © 2004 Elsevier B.V. All rights reserved. doi:10.1016/j.jpowsour.2004.06.041

ponents by controlling the phase content of the partially crystallized glass and enables thermal cycles during stack operation with moderate heating and cooling rates [5]. However, glass-ceramics are disadvantageous for long-term stack operation. Chemical interaction of the sealing glass with the interconnect steel may deteriorate the corrosion resistance of both chromia-forming as well as alumina-forming alloys [4]. The inherent brittleness of glass ceramics and the rigid bonding of the stack components limits the compensation of mechanical and thermal stresses and promotes growth of defects or cracks at the interconnect/glass-ceramic interface. Due to the stiff stack assembly, the reduction of mechanical or thermal stresses by elastic or plastic deformation or by free expansion and contraction of stack components is hampered. Furthermore, a non-destructive dismantling of stacks to replace malfunctioning components is impossible.

To overcome the disadvantages of sealing systems based on glass-ceramics, investigation of alternative sealing concepts has been started. Gosh et al. [6–8] proposed a ceramic sheet as a sealing component consisting of randomly orientated fibres of various oxides, e.g.  $Al_2O_3$ ,  $ZrO_2$  or  $TiO_2$ . The interspace between the fibres was filled with spheroidal ceramic particles of different sizes, and an organic binder was added. A pre-compression of the compound down to approximately 0.2 mm was found to be necessary before use. For the same purpose, use of an  $Al_2O_3$  fleece is reported [9]. The elastic sealing component tested has a quadratic geometry and a thickness of 0.3 mm. Along the sealing planes of a stack a leakage rate of less than 2% of the total gas flow was measured.

Compressive single layer or multi-layer seals have been tested in a few studies. Besides the use of metallic gaskets, e.g. as sealants in a single-cell test bed [10], systematic investigations [11,12] reveal the sealing effect of muscovite and phlogopite mica in the form of paper or single crystal sheets with a thickness of 0.1 mm. The material was tested under a load of approximately 0.7 MPa as plain mica or with a glass-ceramic interlayer between the mica and the material to be joined, a so-called hybrid design. It was noticed that the leakage rate during thermal cycling at 800 °C decreases with a better TEC adaptation of the seal and the adjacent stack components. The planar orientation of single crystal

Table 1

Starting materials	used in	this	investigation
--------------------	---------	------	---------------

mica platelets provides a sealing effect that is superior to the three-dimensional structure of paper containing flakes. Novel investigations deal with the combination of mica multilayer compressive seals with two thin silver layers instead of the abovementioned glass-ceramic [13]. The Ag/mica showed good thermal cycle stability and improved leakage behavior compared to the corresponding mica seals with glass interlayers, although the leakage rates were enhanced.

Preliminary tests with commercial sealing components were conducted at the Institute of Materials and Processes (IWV) of Forschungszentrum Jülich. Since they did not show a load-deflection behavior suitable for SOFC application, metallic gaskets tailored to the planar stack design were developed by an embossing technique. The latter is a feasible and cost-effective production route [14]. To improve the mechanical behavior and the gas tightness, the wave crests were filled with mica powder to which an organic binder was added. The dependence of the mechanical properties and the leakage behavior of these compressive seals on the filling material as well as the relationship between the TEC of the adjacent materials and the leak rate were investigated for different geometries (circular and quadratic). The tests were conducted in contact with the high-temperature-resistant steels and the 8YSZ electrolyte [15]. Aspects of TEC mismatch were discussed together with the surface roughness of the compressed components and the material degradation by chemical interaction in [16].

The present paper focuses on a characterization of alternative SOFC sealing materials such as mica, ceramic fibre sheets, embossed metal foils and combined arrangements of these materials. Creep and deformation behavior is addressed. In addition, the leakage rates are measured as a function of compressive load in a test rig. A sandwich arrangement of mica paper and embossed metal foils is described with which the leakage rate can be clearly reduced. Finally, first results of a stack leakage test are described.

#### 2. Experimental

The data of all materials used in this investigation are summarized in Table 1. Different sealing concepts based on these

Component	Material	Trade name and manufacturer	Further processing
Metal sheet	FeCrAlY-alloy 1.4767 thickness 0.2 mm	Aluchrom Y Hf ThyssenKrupp, Germany	Embossing with 80 kN, laser cutting
Mica powder	Phlogopite powder $d_{10} = 12 \ \mu m \ d_{50}$ = 44 \ \mu m \ d_{90} = 82 \ \mu m	Suzorite 200 S Suzorite Mica Prod. Inc., Canada	Mixing with organic binder, filling the paste in the wave crests
Mica paper	Natural mica with binder thickness 1 mm	Statotherm-HT Burgmann, Germany	Laser cutting
Mica paper with metallic inlay	Vermiculite mica with binder and metallic inlay (1.4401) thickness 1 mm	Thermiculite 815 Flexitallic, UK	Laser cutting
Ceramic paper	$Al_2O_3$ -SiO <sub>2</sub> fibres Ø 2.0–3.0 µm thickness 2 mm	FibrePap Schupp Industrie-keramik, Germany	Conventional cutting



Fig. 1. Schematic drawings of different compressive sealing concepts: (a) corrugated metal sheet with filled with mica paste; (b) sandwich arrangement of mica paper and metal sheet; (c) plain mica paper.

materials were tested (Fig. 1). As already reported elsewhere [14], metallic profiles with embossed wave crests are easy to manufacture. To reduce their inherent affinity to creep under SOFC conditions, the wave crest was filled with mica powder (Fig. 1a). A further improvement of creep resistance was expected by replacing the loose powder particles with a flat compressive inlay made of mica paper. Its enhanced elasticity is achieved by the layerwise arrangement of single mica platelets. The geometry of the composite seal had to be adapted to the flat inlay (Fig. 1b). To improve gas tightness, the embossed top sheet and the flat bottom sheet were welded by laser. For comparison, plain mica paper and plain ceramic paper, as another candidate filler material, were applied (Fig. 1c). Fig. 2 shows the microstructure of both filler materials.

## 2.1. Compression test of sealing components

To study the deformation behavior of the sealing concepts at room temperature and SOFC operating temperature ( $800 \,^{\circ}$ C), rectangular sheets (25mm × 25mm) of each sealing material were prepared. The specimens were loaded under compression in an INSTRON 1362 high-temperature-testing machine. Starting the test with a pre-load of 20 N, the samples were compressed at 50 N/min to 900 N (1.4 MPa) and subsequently unloaded at 200 N/min. The loading-unloading cycle was repeated at least once, in some cases up to 4 times. Residual stresses partially influenced the determination of the

irreversible plastic deformation. In the case of mica paper, an elastic spring-back after unloading was observed. Therefore, the elastic recovery was determined between the maximum applied load of 900 N and an abitrarily defined low load of 50 N. The irreversible plastic deformation was estimated from a load of 20 N. With respect to the influence of temperature on the deformation behavior, all tests were carried out at room temperature and at SOFC operating temperature ( $800 \,^{\circ}$ C) in air. In the high-temperature test the samples were heated up to  $800 \,^{\circ}$ C at a rate of 8 K/min. The creep behavior of the sealing materials was monitored under a force of 900 N at  $800 \,^{\circ}$ C at a compression load rate of 50 N/min. This compression load was maintained for at least 1.5 h, during this period the deformation behavior was recorded. Thereafter the samples were unloaded at 200 N/min.

## 2.2. Leakage test

The measurement of load and related leakage was carried out simultaneously in a simple test facility [14,15]. Square seals with an outer geometry of 46mm  $\times$  46 mm were tested. All gaskets were placed between two faces of a mould, machined from the interconnect steel 1.4742. An additional bottom part of the mould was machined to mount an SOFC cell with a size of 50mm  $\times$  50 mm. This assembly was used to simulate the sealing between interconnect and electrolyte of the SOFC. The complete device was mounted in a furnace and loaded by hydraulic pistons. The test arrangement



Fig. 2. Microstructure of filler materials: (a) mica paper (Thermiculite 815); (b) ceramic paper (FibrePap).



Fig. 3. Testing device for square model geometries.

is shown in Fig. 3. All experiments were conducted under load control (maximum load 10 kN). An absolute internal pressure of 1500 mbar (0.15 MPa) was built up with argon gas. This value exceeds the pressure under real SOFC stack conditions, where an absolute pressure between 1050 and 1200 mbar is commonly used. The leakage was determined by a conventional flow meter. The detection limit of the flow meter was 1 ml/min, which corresponds to a leakage rate of  $<1 \times 10^{-4}$  mbar l/s mm for the given pressure and a sealing length of 184 mm. To simplify the comparison between sealing loads of different sealing concepts, a standardized load related to the sealing length is given (unit N/mm). For flat seals, also the area load is considered (unit MPa).

All compressive seals were examined following the standardized test performance. After mounting the compression rig with the specimen in the tube furnace a pre-load force of F = 0.12 kN and an absolute pressure of p = 1500 mbar (0.15 MPa) was applied. Then the furnace was heated up to 800 °C at a heating rate of 10 K/min. After a holding time of 1 h an increase of the compression load in steps of 0.5 kN was repeated until the gas flow <1 ml/min was detected. The test device was maintained under these conditions for a period of 15 h. The following unloading procedure was conducted with a stepwise decrement of 0.5 kN until a gas flow >1 ml/min was indicated. Subsequently, the furnace was switched off. The specimen was taken out at room temperature.

#### 2.3. Preliminary stack test

A leakage test bed for planar SOFC stacks was constructed (Fig. 4). It is designed to make quantitative analyses of leakage currents in SOFC stacks between the anode and the cathode gas circuits and between both circuits and the surrounding atmosphere by mass spectroscopy. Therefore, air as the oxidant and varying compositions of Ar/H<sub>2</sub> as the fuel were fed in, controlled by flow rate and temperature. All gases can be mixed with He as an optional tracer gas. Systematic test series are under way characterizing and comparing different sealing concepts for planar SOFC stacks by mass spectroscopy. To accomplish this, the gas compositions before and after both gas circuits were measured separately at four independent control points. A general pre-condition for the quantitative evaluation of leakage currents is a sufficient gas tightness of the complete test bed including all valves and measuring devices. In addition, a small gas flow from the different measurement sites of the gas mains to the mass spectrometer has to be assured. This provides constant stack operating conditions during the measurement of the gas compositions. At the gas inlet gas flow is limited to 1201/h at a maximum temperature of about 100 °C (fuel) and an absolute pressure of 1600 mbar. The test bed is connected to a clamshell furnace housing the stack, which can be heated up to 1100 °C. The stack is loaded up to 1500 kg by a level system outside the furnace to determine the influence of the applied load on the gas tightness. At the moment, the test rig is not designed for electrochemical testing of the stack.

Considering the results of the deformation and leakage tests, a stack based on the Jülich standard design was modified for the application of compressive seals. Fig. 5 shows where the sealings were located in this stack. The flat seal made of the embossed metallic sheet combined with mica paper as



Fig. 4. Testing device for planar SOFC stacks.



Fig. 5. Position of seals in the modified Jülich design. Seal 1: sandwich arrangement metallic sheets with mica paper, seal 2: plain mica paper.

inlay was used to separate the anode and cathode gas circuits (Fig. 5,  $\rightarrow$  seal 1). For sealing the air manifold ( $\rightarrow$  seal 2) plain mica paper (Thermiculite 815) was chosen, which was cut by laser to the final dimensions. Before mounting the stack, the mica paper was precompacted to a height of 0.8 mm to avoid excessive deformation during stack loading. The application of metallic seals was restricted to seal 2 to avoid short circuits between adjacent interconnects.

The stack was mounted in the test rig and then loaded with 400 kg (~0.8 MPa relative to the area of the flat seals). The stack was heated to 790 °C at 10 K/min. Then, air (20.9 vol.% O<sub>2</sub>, 78.1 vol.% N<sub>2</sub>, 0.9 vol.% Ar) was fed through the cathode gas circuit. For the anode circuit, 96 vol.% Ar with 4 vol.% H<sub>2</sub> was used. To achieve a quantitative analysis of the leakage, the fuel gas was doped with helium resulting in the gas composition of 48.9 vol.% Ar, 2.0 vol.% H<sub>2</sub>, 49.1 vol.% He. The stack was operated for 11 days under constant conditions afterwards the load was varied to 50 kg (~0.1 MPa), again 400 kg ( $\sim$ 0.8 MPa) and at least 1490 kg ( $\sim$ 2.9 MPa) while keeping the gas flow constant. To determine the leakage current, the gas composition of the fuel gas outlet was analysed. After cooling down to room temperature, the stack was unloaded and then demounted for post-operational characterization.

### 3. Results

#### 3.1. Compression behavior of sealing components

The load-deflection curves of mica paper with metallic inlay at room temperature demonstrate the characteristic behavior of compressive seals (Fig. 6). The strong irreversible plastic deformation during the first cycle is mainly caused by flattening of the specimen, followed by comparatively small elastic recovery (Table 2). The plastic deformation during further cycles decreases, but the elasticity seems to stay almost constant during unloading. Only a slight reduction of the elastic recovery of about  $1-2 \mu m$  per cycle was observed. Considering the high plastic deformation during the first cy-



Fig. 6. Loading-unloading cycles of mica paper with metallic inlay (Thermiculite  $815^{TM}$ ) at room temperature.

cle a pre-compression of the seals before application in a stack is advised.

Creep of the metals in the sealing composites is observed during the experiments under constant load at SOFC operating temperature (800 °C). Regarding the limited test period of maximum 24 h, only an estimation of the deformation potential is given. Corresponding to the load-deflection curves, the corrugated profile of the ferritic steel is more susceptible to creep. An irreversible plastic deformation of about 50% of the initial height was already measured after 3 h (Fig. 7a, curve I). Mica powder as a filler of the corrugations clearly reduces the creep without preventing it (Fig. 7a, curve II). In contrast to the metallic profiles, the creep of mica paper almost saturates at a constant value after 1 h (Fig. 7b). In the presence of a metallic inlay the creep resistance of mica paper is reduced (Fig. 7c), and even after 20 h progressing creep is observed.

Equally important for practical application is the elastic recovery during unloading. The corrugated profile made of ferritic Aluchrom Y Hf<sup>TM</sup> alloy (height 1 mm) shows an elastic recovery of approximately 2  $\mu$ m at 800 °C (Fig. 8a), compared to ~19  $\mu$ m at room temperature. As profile filler, mica powder slightly improves the elastic effect up to ~8  $\mu$ m (not shown here). The elastic behavior of mica paper is by far superior to metallic seals. This material has an even higher recovery at SOFC operating temperature (~57  $\mu$ m, Fig. 8b) than at room temperature (~40  $\mu$ m). The decrease in the elasticity of mica paper with metallic inlay was expected from the proceeding creep under constant load (Fig. 7c). There-

Table 2

Plastic deformation and elastic recovery of mica paper with metallic inlay depending on the number of loading-unloading cycles at room temperature

	Plastic deformation (µm)	Elastic recovery (µm)
1st cycle	225	29.8
2nd cycle	37	27.8
3rd cycle	14	26.8
4th cycle	14	26.4
5th cycle	13	25.4
-	Σ303	



Fig. 7. Creep deformation of sealing materials at 800  $^{\circ}$ C (a) I: corrugated profile, made of ferritic steel Aluchrom Y Hf<sup>TM</sup>, II: corrugated profile, filled with mica; (b) mica paper (Statotherm); (c) mica paper with metallic inlay (Thermiculite 815).

fore, the elastic recovery was reduced to  $\sim 30 \,\mu m$  (Fig. 8c). Nevertheless, this type of mica paper was preferred for the stack test due to the fact that metallic inlays remarkably enhance the stability of mica papers because flaking of mica particles is prevented during long-term operation. Ceramic papers were found to be less attractive for SOFC applications because of their high irreversible deformation (>50%) combined with comparatively low elasticity (elastic recovery  $\sim 20 \,\mu m$  at a starting height of 2 mm).

## 3.2. Leakage tests

Fig. 9 shows the load-dependent leak rate of a corrugated profile made of Aluchrom Y Hf filled with mica powder. At



Fig. 8. Deformation and elastic recovery of sealing materials at  $800 \,^{\circ}$ C: (a) corrugated profile, made of ferritic steel Aluchrom Y Hf, height 1 mm; (b) mica paper (Statotherm, thickness 1 mm); (c) mica paper with metallic inlay (Thermiculite 815, thickness 1 mm).

16.3 N/mm the flow rate drops below the detection limit corresponding to a leak rate  $<1 \times 10^{-4}$  mbar l/s mm. After 16 h, the reduction of the load to 2.7 N/mm shows no increase of the leakage. The rate only increases after complete unloading. This favorable leakage behavior might be attributed to similar material properties of the adjacent components or the development of interfacial layers. A further load-dependent leakage test was conducted with the sealing component in contact with the electrolyte. No damage occurred to the SOFC under the test load scheme.



Fig. 9. Sealing behavior of corrugated profile (Aluchrom Y Hf), filled with mica powder, sealing between 1.4742 and electrolyte of SOFC at 800 °C.



Fig. 10. Sealing behavior of mica paper with metallic inlay (Thermiculite 815) at 800  $^\circ\text{C}.$ 

In the leakage tests with mica paper higher sealing loads up to 54 N/mm ( $\sim$ 15 MPa) had to be applied, but the impact of gas tightness was very small (Fig. 10). A clear improvement of the sealing behavior was achieved when the mica paper was sandwiched between metallic sheets (Fig. 11). Using this sandwich arrangement the detection limit was already reached at a compression load of 2.7 N/mm ( $\sim$ 0.7 MPa). Al-



Fig. 11. Sealing behavior of composite seal, made of a flat metallic sheet (Aluchrom Y Hf) supported by mica paper at 800  $^{\circ}$ C.



Fig. 12. Thermal cycling of corrugated profile (Aluchrom Y Hf) between 800  $^{\circ}$ C and room temperature under constant load of 3 kN, profile filled with mica powder, sealing between 1.4742 and electrolyte of SOFC.

though the number of leakage tests is still limited and further reproduction experiments are required, the results indicate the promising potential of the sandwich arrangement. Gas-tight sealing of SOFC stacks at acceptable loads was expected.

Despite the results of the test with permanent load, gas leakage was observed during thermal cycling of corrugated profiles filled with mica powder (Fig. 12). TEC deviations probably cause the leak rates by shear displacement along the contact areas. In the present investigation, the TEC of the testing tool (steel 1.4742) is  $\alpha_{0-800 \circ C} = 12.5 \times 10^{-6} \text{ K}^{-1}$ , of the corrugated seal (Aluchrom Y Hf)  $\alpha_{0-800 \circ C} = 14.3 \times 10^{-6} \text{ K}^{-1}$  and of the electrolyte of SOFC (8 YSZ)  $\alpha_{0-800 \circ C} = 10.5 \times 10^{-6} \text{ K}^{-1}$ . Thus, the matching of TECs seems to be very important. For further attempts, the application of metallic sheets made of the interconnect steel is advisable to reduce the TEC mismatch.

### 3.3. Preliminary stack test

Fig. 13 shows the stack after post-operational demounting. The mica paper used for the sealing of the air manifold adhered strongly to the interconnects leading to destruction of the seal. Therefore, a complete recycling of all stack components does not appear to be possible with this design. The anode and cathode side of the SOFC after operation are shown in Fig. 14. Obviously, it was not possible to reduce the NiO of the anode cermet to metallic Ni under the given conditions. Only a slight color change was found near the fuel gas inlet (Fig. 14a). The reason for this behavior is thought to be the high leakage rate of the plain mica paper used for sealing the air manifold. As indicated by preliminary experiments, even the maximum stack loading of 1490 kg (~2.9 MPa) was not enough to ensure sufficient gas tightness. As a consequence, the amount of H<sub>2</sub> in the fuel gas was completely oxidized immediately after flowing into the anode circuit. This assumption was supported by data from mass spectroscopy on the fuel gas outlet (Table 3), where H<sub>2</sub> was no longer detected. The gas composition at the fuel outlet also indicated that a leakage flow of approximately 10% from the air to



Fig. 13. Stack with compressive seals after testing its gas tigthness.



Fig. 14. SOFC after testing in a stack with compressive seals: (a) anode side; (b) cathode side.

Table 3
Composition of fuel gas before injection and after passing through the stack
with compressive seals

	Fuel gas inlet (vol.%)	Fuel gas outlet (vol.%)
Ar	48.9	51.0
He	49.1	37.0
H <sub>2</sub>	2.0	_
$N_2$	_	10.4

the fuel gas manifold had taken place. This result was only marginally influenced by varying the stack loading in the range of 50-1490 kg.

As a promising result, the SOFC was able to withstand even the highest load without damage. Under this load, a severe plastic deformation of the compressive seals was found, reducing the height of the mica paper from 0.8 to 0.6 mm. Consequently, clearly pronounced fingerprints of the Ni mesh on the anode side (Fig. 14a) as well as the gas channels on the cathode side (Fig. 14b) indicates an enhanced electrical contact compared to conventional stacks sealed with rigid glass ceramics. Additional electrochemical tests are necessary to demonstrate this improvement. However, in order to perform these tests the leakage rate of the mica paper must be reduced, for example by introducing a soldering glass or metal as proposed by PNNL [11–13]. Corresponding attempts are under way.

## 4. Conclusions

Compressive seals with a metal-mica arrangement provide a high potential for SOFC application avoiding the inherent drawbacks of glass-ceramics. For that purpose metallic seals, mica paper and combined arrangements of these materials were investigated under SOFC operating conditions.

Corrugated metallic seals achieving a leak rate of  $<1 \times 10^{-4}$  mbar l/s mm at an adequate compression force of 16 N/mm seem to be promising for SOFC requirements. The insufficient spring-back effect of <0.2% and their strong susceptibility to creep require the use of appropriate filler materials.

Plain mica paper exhibits the highest elastic recovery of about 5.8%, but a detectable improvement in gas tightness does not take place before a very high level of compression load of more than 50 N/mm (15 MPa) is applied. Mica paper is less stable under high temperature conditions after the binder has been burnt off due to the flake-like structure of the material. This disadvantage is compensated by the use of commercial mica paper with a thin metallic inlay, which improves the durability during long-term operation.

A sandwich arrangement of mica sheet embedded between embossed metallic profiles was found to be the most suitable combination. At a compression load of 2.7 N/mm (0.7 MPa) the flow rate dropped below the detection limit of the testing device. However, it has to be considered that this seal is not suitable for sealing the manifolds between adjacent interconnects due to the occurrence of short circuits.

Leakage investigations on complete SOFC stacks were performed in a test bed where leak rates in and between the gas circuits of the anode and cathode side can be analyzed by differential mass spectroscopy. A planar stack based on the Jülich standard design was modified for the application of compressive seals. While the air manifold was sealed with plain mica paper, a composite seal consisting of a sandwich arrangement of a mica paper between embossed metallic sheets was used between the interconnect and the SOFC itself. As already indicated by preliminary leakage tests, the gas tightness of the plain mica paper was not sufficient for successful stack operation. Nevertheless, the compressibility of the seals combined with the ability of the SOFC to withstand loads on the stack of up to 2.5 MPa without failure was found to be promising by improving the electrical contact compared to conventional stacks sealed with rigid

glass-ceramics. Attempts to optimize the presented sealing concept are under way.

### References

- T. Schwickert, P. Geasee, A. Janke, R. Conradt, U. Diekmann, Electrically insulating high-temperature joints for ferritic chromium steel, in: Proceedings of the IBSC 2000, Albuquerque, NM, USA, 2000, pp. 116–122.
- [2] K.L. Ley, M. Krumpelt, R. Kumar, J.H. Meiser, I. Bloom, Glassceramic sealants for solid oxide fuel cells: part I physical properties, J. Mater. Res. 11 (1996) 1489–1493.
- [3] N. Lahl, K. Singh, L. Singheiser, K. Hilpert, D. Bahadur, Crystallisation kinetics in AO-Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub>-B<sub>2</sub>O<sub>3</sub> glasses (A = Ba, Ca, Mg), J. Mater. Sci. 35 (2000) 3089–3096.
- [4] Z. Yang, J.W. Stevenson, K.D. Meinhardt, Chemical interactions of barium–calcium–alumosilicate-based sealing glasses with oxidation resistant alloys, Solid State Ionics 160 (2003) 213–225.
- [5] L. Blum, L.G.J. de Haart, I. Vinke, D. Stolten, H.P. Buchkremer, F. Tietz, G. Blaß, D. Stöver, J. Remmel, A. Cramer, R. Sievering, Planar anode substrate type SOFC kW-class stack development, in: J. Huijsmans (Ed.), Proceedings of the fifth European Solid Oxide Fuel Cell Forum, 1–5 July 2002, Lucerne, Switzerland, 2002, pp. 784–790.
- [6] WO 02/17416 A2, 2002.
- [7] D. Gosh, R. Wang, E. Brule, P. Tang, Performance of anode supported planar SOFC cells, solid oxide fuel cell VI, in: S. Singhal, M. Dokiya (Eds.), Electrochemical Society Proceedings, 1999, pp. 822–829.
- [8] D. Gosh, M.E. Pastula, R. Boersma, D. Prediger, M. Perry, A. Hovath, J. Devitt, Development of low temperature SOFC systems for remote power and home cogen applications, in: Proceedings of the Fuel Cell Seminar, Washington, D.C., 2000, pp. 511–514.
- [9] DE 4016157 A1, 1990.
- [10] J. Kim, A. Virkar, The effect of anode thickness on the performance of anode-supported solid oxide fuel cells, solid oxide fuel cell VI, in: S. Singhal, M. Dokiya (Eds.), Electrochemical Society Proceedings, 1999, pp. 830–839.
- [11] S.P. Simner, J.W. Stevenson, Compressive mica seals for SOFC applications, J. Power Sources 102 (2001) 310–316.
- [12] Y-S. Chou, J.W. Stevenson, L.A. Chick, Ultra-low leak rate hybrid compressive mica seals for solid oxide fuel cells, J. Power Sources 112 (2002) 130–136.
- [13] Y.-S. Chou, J.W. Stevenson, Novel silver/mica multilayer compressive seals for solid oxide fuel cells: the effect of thermal cycling and material degradation on leak behavior, J. Mater. Res. 18 (2003) 2243–2250.
- [14] M. Bram, S.E. Brünings, F. Meschke, W.A. Meulenberg, H.P. Buchkremer, R.W. Steinbrech, D. Stöver, Application of metallic gaskets in SOFC stacks, in: H. Yokokawa, S.C. Singhal (Eds.), Proceedings of the seventh International Conference on Solid Oxide Fuel Cells, SOFC VII, 3–8 June 2001, Tsukuba, Japan, 2001, pp. 875–884.
- [15] M. Bram, S. Reckers, P. Drinovac, S.E. Brünings, R.W. Steinbrech, H.P. Buchkremer, D. Stöver, Basic investigations on metallic and composite gaskets for an application in SOFC stacks, in: J. Huijsmans (Ed.), Proceedings of the fifth European Solid Oxide Fuel Cell Forum, 1–5 July 2002, Lucerne, Switzerland, 2002, pp. 847–854.
- [16] S. Reckers, M. Bram, H.P. Buchkremer, D. Stöver, Leakage investigations of compressive metallic seals in SOFC stacks, in: J. Huijsmans (Ed.), Proceedings of the fifth European Solid Oxide Fuel Cell Forum, 1–5 July 2002, Lucerne, Switzerland, 2002, pp. 847–854.