Final Report

Metal supported SOFC technology for Stationary and Mobile Applications (METSAPP)

(ForskEL project No. 2012-1-10806)

Report prepared by
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Final report

1.1 Project details

Project title	Metal Supported SOFC Technology for Stationary and Mobile Applications (METSAPP)
Project identification (program abbrev. and file)	Energinet.dk project no. 2012-1-10806
Name of the programme which has funded the project	ForskEL
Project managing company/institution (name and address)	Department of Energy Conversion and Storage, Technical University of Denmark, Frederiksborgvej 399, DK-4000, Roskilde
Project partners	Topsoe Fuel Cells A/S (TOFC)
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1.2 Short description of project objective and results

The main objective of the METSAPP was to develop the novel cells and stacks based on a robust and reliable up-scalable metal supported technology. During the course of the project novel anode materials that are corrosion resistant along with high performing cathode materials were developed, integrated into the metal supported SOFCs (MS-SOFCs) using the environmentally friendly manufacturing process. The cell manufacturing process is up-scalable and flexible and this was demonstrated through the fabrication of more than 500 MS-SOFCs in different sizes (up to 300 cm^2). Testing of MS-SOFCs with the novel anodes showed significant improvement in their robustness and corrosion resistance. Electrochemical testing showed a maximum performance of ca. 0.32 Ω-cm^2 and a durability of 1-3% degradation for 1000 h. These results are highly promising with further scope for improvement. Furthermore, these MS-SOFCs were integrated in to the SOFC stacks using different stack designs and tested.

Det vigtigste mål i METSAPP var at udvikle nye celler og stakke baseret på en robust og pålidelig metalbåren teknologi der kan opskaleres til industriel skala. I løbet af projektet har man udviklet nye anodematerialer der ikke korroderer, samt højtydende katodematerialer. Materialerne er blevet integreret i metalbårne SOFC-celler vha. en miljøvenlig fremstillingsproces. Det er blevet demonstreret at fremstillingsprocessen er fleksibel og kan opskaleres, og der er blevet fremstillet flere end 500 celler i forskellige størrelser (up to 300 cm^2). Test af cellernes mekaniske og korrosionsbestandige egenskaber viste væsentlige forbedringer. Elektrokemiske test viste en maksimal ydelse på ca. 0.32 Ω-cm^2 og en degradering på 1-3% pr. 1000 timer. Selv om degraderingsraten stadig skal forbedres, er dette meget lovende resultater. Endelig blev brændselscellerne integreret i stakke i forskellige design og testet.

1.3 Executive summary

State of the art SOFC technology for stationary and transportation applications employs either planar or tubular ceramic anode-supported or electrolyte-supported SOFC cells. However, the SOFC technology faces many hurdles for commercialization, because cost reduction, reliability and extended lifetime are required. In order to improve durability and cost efficiency of the cells, stacks, and system, much of the development has focused on lower operation temperature, increased power density, and material savings. In the conventional anode-supported cell design, the mechanical support is a brittle ceramic or cermet, and contains expensive materials. In contrast, the metal-supported solid oxide fuel cell (MS-SOFC) design utilizes ceramic layers only as thick as necessary for electrochemical function; the mechanical support was made from inexpensive and robust porous metal, and the electrochemically active layers were applied directly to the metal support. Due to their specific material properties, metal-supported cells offer the possibility to control mechanical stresses and thereby improve reliability and durability by design. The metal supported cell stacks become much more cost effective because of much lower safety factors are needed in the design using metallic materials. Moreover, the novel metal-supported concept avoids use of large amounts of potentially problematic Ni contents by replacing it with more environmentally friendly and cost effective ferritic stainless steel.

In the previous FCH JU project, METSOFC, in which the development of metal supported cells was significantly advanced, the main challenges in improving the robustness and performance of MS-SOFCs were identified as:

- Severe corrosion of the metal particles in anode backbone layer
- Development of novel anode backbone designs
- Delamination of the cathode layer during thermal cycling
- Integration of the novel cell components into cells
- High volume manufacturing of the cells for stacks

Therefore, the main objective of the METSAPP project is to develop novel low cost, durable, robust and reliable metal based stacks. The project is organized along the following lines: cell components development, integration and manufacturing in WP2, evaluation of the electrochemical, corrosion and mechanical performance of the cells and stacks in WP3, modelling of performance, reliability and durability in WP4, stack designs development, stack building and stack testing in WP5, Protective coatings for interconnects in WP6, dissemination and exploitation of the results in WP7 and consortium dissemination in WP8. All these activities are aimed at developing the robust metal supported cells and stacks through cost-efficient and scalable techniques. The Danish partners, DTU and TOFC are heavily involved in WP2, WP3 and WP5 and contributed to other WPs in small extent.

Based on the results and identified challenges from the previous FCH JU METSOFC project, the robust cell development and cell fabrication is addressed through the development and integration of oxidation resistant alloys, protective coating for anode backbone, alternative anode materials and high performance cathode materials. Potential oxidation resistant alloys such as 316L, IN718 and F75 are investigated as a replacement for the FeCr ferritic alloys that is being used in the project as both metal support and anode backbone. Due to the difference in their sintering behaviour, practical restrictions such as availability with required physical characteristics and cost, these alloys are found to be unsuitable with the current cell manufacturing process. Modification of their physical characteristics and addition of other alloying elements to tune their sintering behaviour might render them suitable as alternative alloys. Due to these limitations, FeCr ferritic alloys are used throughout the project for developing other components of the cell.

Different coating materials such reactive element, chromites, titanates, metal oxides and spinels are investigated for their efficiency in improving the oxidation resistance of the FeCr-YSZ based anode backbones in relevant conditions. The titanate based coatings such as doped $SrTiO_3$ and doped TiO_2 were either difficult form at low temperature or unstable in MS-SOFC operating conditions. Metal oxide coatings such CuO were successfully formed at lower temperatures (< 400ºC), however, the CuO was reduced to Cu and the coating was disintegrated into discrete particles in the fuel electrode environment. This disintegration of the coating resulted in an inefficient protection of FeCr surfaces. The other coating materials i.e., reactive elements, chromites and spinels were the most promising. The major challenge with these materials was obtaining the continuous and uniform coating on the FeCr porous structures. The quality of the obtained coatings was strongly dependent on the metal surface quality, impurities, wettability of the coating precursor solutions. The adherence and uniformity of these coatings required further optimization to completely realize their potential in improving the oxidation resistance of metal surfaces. It is realized that both the development of oxidation resistant alloys and protective coatings development require large amount of research and development before they can be integrated into MS-SOFCs. The time and resources available in the project limited the activities and attention is focused on the development of alternative anode backbone materials that showed huge promise.

The alternative anode designs investigated are based on electronically conducting ceramic materials such as doped $SrTiO_3$. Among the doped $SrTiO_3$ materials, DTU focused on the Nb-doped $SrTiO_3$ (STN) materials development and the EU partner USTAN focused on the $(La,Sr)(Ti,Fe,Ni)O_3$ (LSFNT) materials development. Anode designs based on both the titanates showed huge improvement in the oxidation resistance of FeCr particles compared to FeCr-YSZ anode backbones in MS-SOFC operating conditions at both short-term and long-term. The titanate phase appeared to be covering the FeCr metal particles and preventing from the oxidation. The electrochemical performance of the FeCr-STN based anode designs with NiCGO electrocatalyst was encouraging (ASR: 0.54-1.23 Ωcm^2); however, it was not reproducible due to the inconsistency in the cell quality. The integration of STN anode designs was challenging due to the different sintering behaviour in cell processing conditions, resulting in poor adhesion with electrolyte and leaky electrolyte layer. LSFNT materials possess good electrical transport properties and favourable sintering behaviour to be matched with other cell materials, making their integration into the MS-SOFCs successful. The FeCr-LSFNT based anode backbones showed the most promising microstructure with significantly improved adhesion with electrolyte and metal support. Both the short-term and long-term electrochemical testing of the MS-SOFCs with FeCr-LSFNT based anode designs with NiCGO electrocatalyst showed significant improvement (ASR: 0.57 ΩCm^2) in the performance and stability (1-3% degradation over 1000 h) at 650°C, advancing the cell development towards project objective, i.e., development of robust metal supported cells. The electrochemical testing of MS-SOFCs with FeCr-YSZ anode designs with RuCGO electrocatalyst showed the best performance of ASR of 0.32 Ωcm^2, however, the stability (ca. 11% over 1000 h) needs further improvement.

The current cell design requires that the cathode needs to be sintered in-situ during the initial start-up used for electrochemical testing. The different cathode materials, such as $(La_{0.6}Sr_{0.4})_{0.99}CoO_3$ (LSC) and $La_{0.58}Sr_{0.4}Co_{0.2}Fe_{0.8}O_3$ (LSCF) were investigated and compared in the temperature range of 650 – 950 ºC. In contrast to LSCF, the LSC-based cathodes showed excellent sintering capabilities, electronic conductivity and performance. The polarization resistance (R_p) of the LSC based cathodes was 0.05 Ωcm^2, which to our knowledge is the best performance reported in the literature for a low temperature 800ºC in-situ sintered cathodes. The thermal and chemical stability of the LSC based cathodes is verified on single cell in MS-SOFC operating conditions. The same could not be verified on stack level due to the limited stacks availability and testing.

The alternative anode materials and high performance cathode materials are successfully integrated into the MS-SOFCs with different sizes (5 cm x 5 cm, 12 cm x 12 cm and 15.3 cm x 8.3 cm, 14.5 cm x 29 cm). The integration process is optimized to fabricate large number of cells in reduced number of processing steps to demonstrate the scalability of the cell fabrication process. More than 500 cells of different sizes are fabricated during the course of the project indicating the adaptability, scalability and cost-efficiency of the process.

The elastic response of the metal supports indicated that the Young's modulus increased linearly with temperature that could due to the oxide scale formation with increasing temperature. The elastic moduli of the ScYSZ electrolyte is found to have a significant peak around 450 °C, where the stiffness is believed to be lowered due to atomistic reorganizations. This will influence the stress distribution amongst the layers over a temperature cycle, but as it occurs mostly the purely elastic temperature range, little impact is expected. The creep measurements for metal support and interconnect steel showed that in the oxidized state the material behavior became complex. The oxide scale formed from corrosion is a less creeping ceramic coating and thus, an oxide scale minimizes the creep rate. However, if higher stresses are applied, the oxide scale peals off, and the creep rate increases compared to the non-oxidized material. This mechanism also increases the corrosion rate, as the protection from the oxide scale is lost and thus results in further corrosion. Infiltration minimizes the corrosion and thus also the amount of corrosive scale. Therefore, the infiltrated metal support creeps faster. Aging treatment of the infiltrated samples, found to be decreasing the creep rate.

The focus of the stack-related work in WP5 at TOFC is to develop 10-25 cell stacks with optimal compression forces, improved laser welds (of cells to interconnects), improved stack conditioning and glass sealing procedures, capability of internal reforming in the stack, and to eventually test them for at least 1000h. Furthermore, the implementation of a new stack design for metal supported cells is investigated. The cells manufactured in WP2 by DTU are delivered to TOFC. In order to build a successful stack, TOFC explored the sealing methods, e.g., laser welding; cathode sintering methods e.g., in-situ and/or ex-situ sintering, stack composition and conditioning treatments along with mathematical models for stack designing. The work resulted in improvements on all aspects of the stack fabrication steps and conditioning and different modelling & simulation tools for stack design optimization, which lead to the development of promising stacks that are intended for stack testing towards the end of 2014. However, due to the closing of TOFC in August 2014 the stacks were unfortunately never tested. Subsequently, the EU METSAPP consortium identified and included another EU partner, ElringKlinger A/G, to perform the stack related activities in WP5 so that the project objectives are achieved.

Overall evaluation of the project results confirms the feasibility of robust metal supported cells and stacks through cost-efficient and scalable processes using the novel anode designs based on alternative electrode materials that are oxidation resistant and stable at MS-SOFC operating conditions. Reduced number of processing steps in cell fabrication and the fabrication of large number of cells proves the cost-efficiency in cell fabrication. The demonstration of fabrication of larger cells (14.5 cm x 29 cm) indicate their potential for larger SOFC systems for stationary applications. The improved performance and stability exhibited by the alternative anode designs is a crucial step further towards real applications and addressing the most critical issues relevant for commercialization. Further improvements in the durability of the cells facilitate the way for stationary applications.

1.4 Project objectives

1.4.1 Project objectives

The METSAPP project has the principal objective to improve the robustness and lifetime of the metal supported SOFC technology, while simultaneously targeting cost effective up-scaling. The following are the primary objectives of the project.

- Robust metal-supported cell design, $ASR_{cell} < 0.5\ \Omega cm^2$, 650ºC
- Cell optimized and up-scaled to > 300 cm^2 footprint
- Improved durability for stationary applications, degradation < 0.25%/kh
- Modular, up-scaled stack design, stack $ASR_{stack} < 0.6\ \Omega cm^2$, 650ºC
- Robustness of 1-3 kW stack verified
- Cost effectiveness, industrially relevance, up-scale-ability illustrated.

A supplementary objective of the project is to develop partnerships and collaboration with potential and relevant partners interested in development and manufacturing of materials and semi-products required to finalize the total value chain ranging from raw materials to the final SOFC stacks.

1.4.2 Implementation

The work in the project is organized in such a way that the state-of-the-art cells based on FeCr-YSZ anode backbone are manufactured in WP2 in large numbers for various testing activities in WP3 and stack design and stack development activities in WP5. The results from activities in WP3 and WP3 are used for the modelling and simulation activities in WP4. Simultaneously various approaches are used to develop the metal supported cells with improved performance, durability and robustness by working on novel anode materials/design and cathode materials. After integrating the novel components into the cells, qualified cells are forwarded to WP3 for electrochemical performance and oxidation resistance evaluation under relevant conditions. Cells with promising performance are manufactured in required numbers in WP2, and the cells are forwarded to WP5 for their integration in stacks. The project utilized the LEAN spiral concept, avoiding sub-optimization and increasing speed of development and thus efficient interaction between the different work packages (Figure 1).

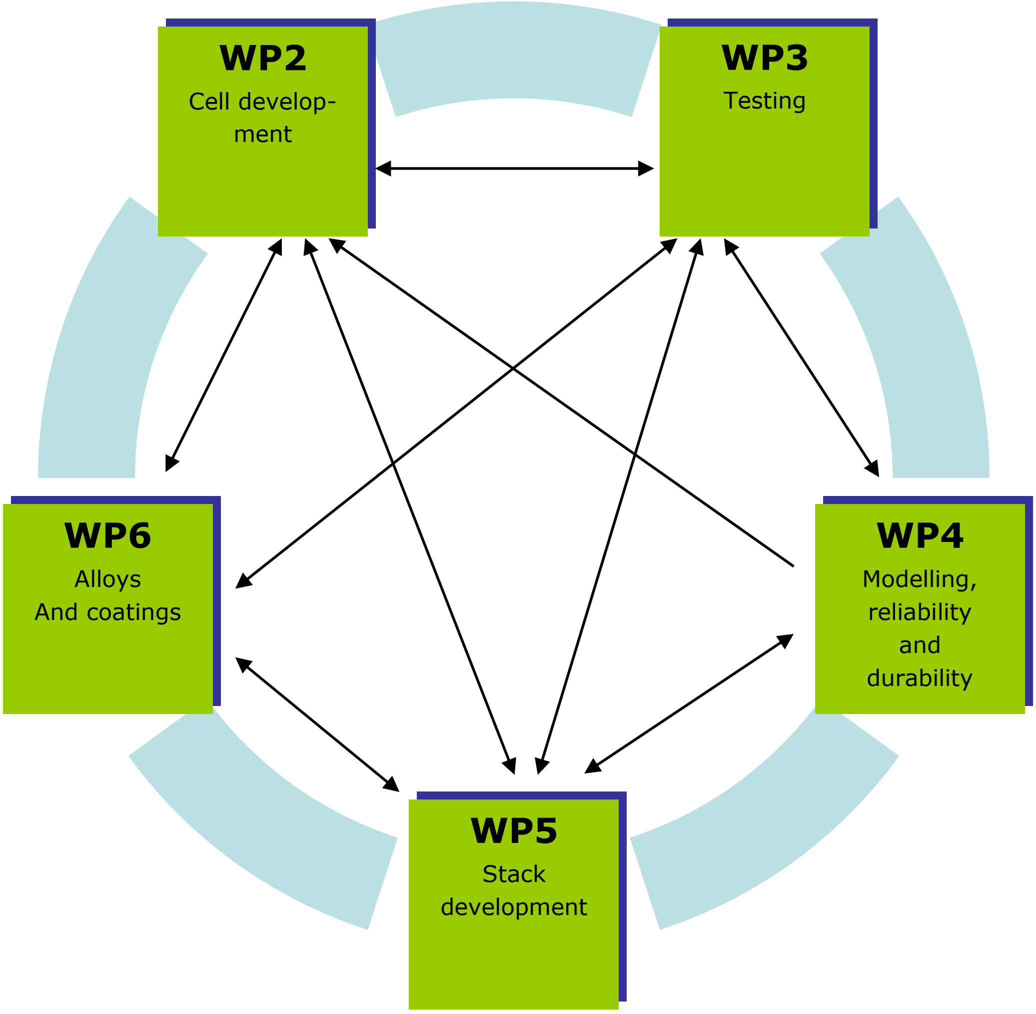

Figure 1: Graphical presentation of work package interaction/interdependence, with effective short cuts (LEAN spiral concept, avoiding sub-optimization and increasing speed of development).

1.4.3 Challenges in implementation

The initial phase of the project is progressed as per the implementation plans; however, in the spring of 2012 major delay of about 6 months in the WP2 activities occurred due to the breakdown of the sintering equipment at DTU. In addition to this, an unexpected deviation is observed in the quality of the raw materials used for cell fabrication. These two challenges delayed the cell development and fabrication activities in WP2 by ca.12 months, which effected the cell delivery to the other WPs in the project. This cumulatively caused delay in the deliverables associated with other WPs. The challenge with the quality of the raw materials is successfully addressed through systematic experimentation and analysis of the raw materials that needed huge number of iteration and time, and large number of cells are manufactured and delivered to TOFC in WP5 for stack development and testing. TOFC successfully managed to integrate these cells into stacks. However, these stacks are not tested as the project encountered another huge challenge in August 2014 in the form of exit of TOFC from the project due to the closure of Topsøe Fuel Cells A/G. Lack of alternative competent organizations for the tasks in WP5 in Denmark forced the EU METSAPP consortium to look for competent organizations out Denmark. ElrignKlinger A/G from Germany is found to be a suitable replacement for TOFC in the EU METSAPP consortium and this whole process delayed the activities by ca. 2 months. Up on reviewing all these delays and change of project partner, an extension of 14 months with modified work plan was applied and the same is approved both by FCH JU and Energinet.dk.

The Danish project partners DTU Energy and TOFC (until August 2014) have participated in WP2-WP5, WP7 and WP8 in close interaction with the European Partners. WP2, part of WP3 and WP5 consist the major activities associated with Danish partners. In the following section, the results mainly focus on the Danish activities and interactions with others partners.

1.5 Project results and dissemination of results

1.5.1 Development of robust anode designs

The metal supported SOFC (MS-SOFC) design is based on a multi-layered structure and can be fabricated by cost effective ceramic processing techniques such as tape casting, lamination, co-sintering and infiltration. This innovative concept includes a half-cell design, where the major material and durability problems associated by the use of Ni-YSZ anodes are circumvented, by the use of an alternative anode structure. The cermet anode backbone was made functional by infiltrating the electro-catalytically active materials. The MS-SOFC concept, details of the different components and the concept of electrocatalyst infiltration can be seen in Figure 2.[1,2,3]

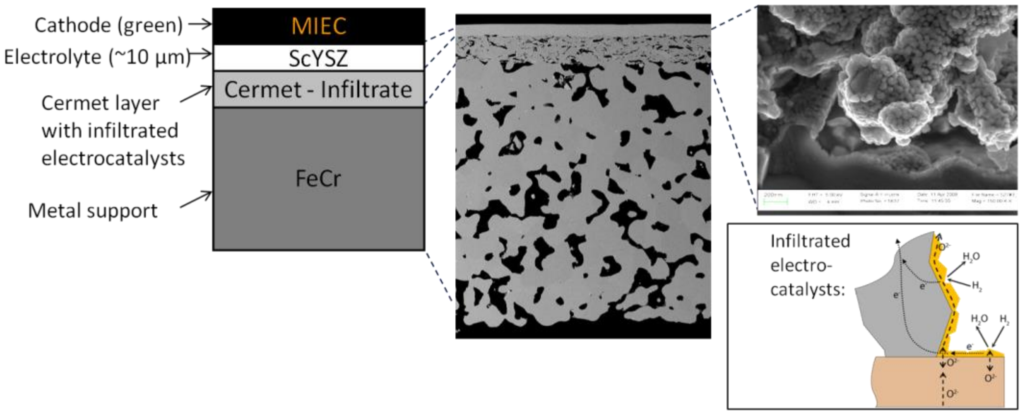

Figure 2: Schematic illustration together with a SEM image showing the concept of the metal supported cell design and electrocatalyst infiltration.

Based on the previous FCH JU METSOFC project results, the main challenges in improving the robustness and performance of MS-SOFCs are identified as:

- Severe corrosion of the metal particles in anode backbone layer
- Development of novel anode backbone designs
- Delamination of the cathode layer during thermal cycling
- Integration of the novel cell components into cells
- High volume manufacturing of cells for stacks.

Oxidation of FeCr metal particles in the FeCr-YSZ anode typically occurs during realistic fuel cell operating conditions (e.g. at high fuel utilization) leading to irreversible cell damage. Despite the positive effects on corrosion inhibition by infiltration of $Ce_{0.8}Gd_{0.2}O_{1.9}$ (CGO), see Figure 3, the corrosion protection of the FeCr particles in the anode layer is not sufficient for long term stability. In order to address this challenge, the design and development of robust anodes is approached through three different concepts. They are listed as below:

- Development of oxidation resistant alloys
- Development of protective nanostructured coatings
- Development of alternative anode designs

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[1] P. Blennow et al., *Fuel Cells,* 11 (2011) 661.

[2] P. Blennow et al., *J. Power Sources,* 196 (2011) 7117.

[3] T. Klemensø et al., *J. Power Sources,* 196 (2011) 9459.

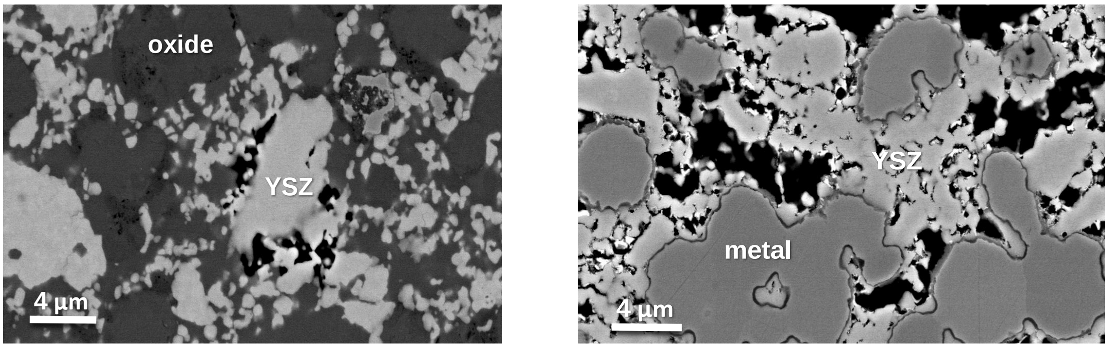

Figure 3: Overview micrographs of FeCr-YSZ cermet without (left) and with (right) CGO infiltration after 2000 h, at 650 ºC, in simulated outlet gas (Ar/H_2/H_2O, $pH_2O/pH_2 = 9$).

1.5.1.1 Development of oxidation resistant alloys

The oxidation resistance of FeCr ferritic alloys, used in the current project, with required particle size is investigated through modelling to estimate their life time (time required for break-away oxidation) at SOFC operating conditions. It is found that the parabolic rate constant (Kp) values around 10^{-16} – 10^{-17} $g^2cm^{-4}s^{-1}$ (or even lower) are needed for lifetimes >10 kh with the particle sizes currently used in the anode layer. These are even harsher requirements compared with other reports in the literature.[4]

Keeping the above requirements in perspective, search is carried out for alloys that can be an alternative to FeCr ferritic alloys. However, the processing conditions such as sintering in low pO_2 at high sintering temperature, imposed by the cell manufacturing process, restricted the number of alternative corrosion resistant alloys. Potentially oxidation resistant alloys such as 316L, IN718 and F75, are investigated for their compatibility with the developed cell manufacturing process. Due to the differences in their sintering behaviour, practical restrictions such as availability with required physical characteristics and cost, these alloys are found to be unsuitable with the current cell manufacturing process. Modification of their physical characteristics and addition of other alloying elements to tune their sintering behaviour might render them suitable as alternative alloys. Due to these limitations, FeCr ferritic alloys are used throughout the project for developing other components of the cell.

1.5.1.2 Development of protective nanostructured coatings

In the second approach for developing robust anodes, various protective coating materials are investigated to evaluate their efficiency in improving the oxidation resistance of the FeCr particles in FeCr-YSZ based anodes. Figure 4 shows a hypothetical schematic of how the protective coatings are intended to work on the FeCr-based materials in the metal-supported SOFC. The primary function of the coating is to limit the formation of Cr_2O_3 on the FeCr particles. The coating is also expected to move the reaction zone away from the FeCr surface to lower the accelerated oxide formation upon drawing a current [5]. The potential coating materials should possess high electronic and low oxygen ion conductivities, prevent Cr-diffusion, compatible with electrocatalyst and demonstrate physical, chemical and microstructural stability. The electrocatalytically active phase infiltrated on top of the protective coating was composed of CGO and minor amount of Ni (10 wt.% with respect to CGO), which is currently the state-of-the-art (SoA) infiltrate used in the MS-SOFC.[1,2,3,5]

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[4] M.C. Tucker, *J. Power Sources,* 195 (2010) 4570.

[5] J. Nielsen, T. Klemensø, P. Blennow, *J.Power Sources,* 219 (2012) 305.

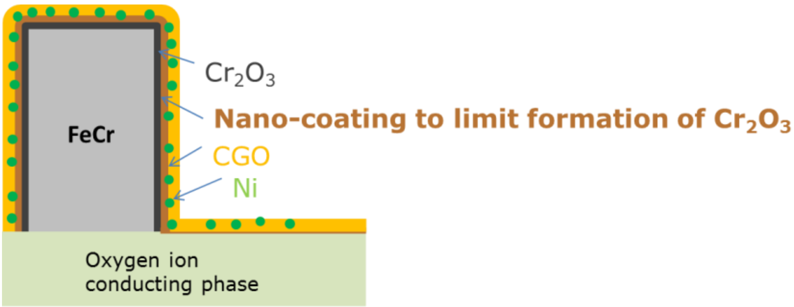

Figure 4: Schematic illustration showing how the nano-structured coatings are intended to inhibit the formation of Cr_2O_3 on the FeCr-based material in the metal-supported SOFC.

Different coating materials such as reactive elements, chromites, titanates, metal oxides and spinels are investigated for their efficiency in improving the oxidation resistance in relevant conditions. The titanate based coatings such as doped $SrTiO_3$ and doped TiO_2 were either difficult form at low temperature or unstable in MS-SOFC operating conditions. Metal oxide coatings such CuO were successfully formed at lower temperatures (< 400°C), however, the CuO was reduced to Cu and the coating was disintegrated into discrete particles in the fuel electrode environment (Figure 5). This disintegration of the coating resulted in an inefficient protection of FeCr surface.

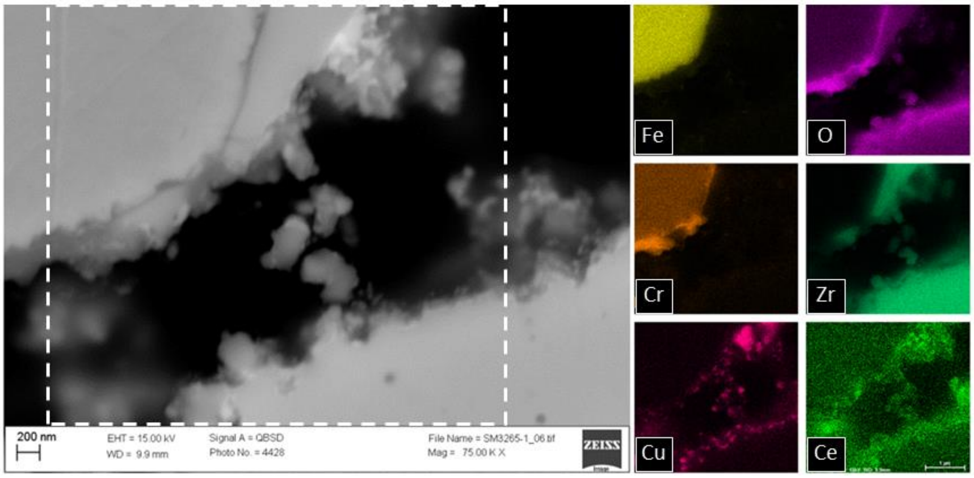

Figure 5: SEM and EDS (part of the anode layer) of CuO + Ni:CGO infiltrated MS-SOFC showing the presence of Cu as nano-sized particles after electrochemical characterization. The dashed area represents the area where EDS is acquired.

The other coating materials i.e., reactive elements, chromites and spinels were the most promising. The major challenge with these materials was obtaining the continuous and uniform coating on the FeCr porous structures. The quality of the obtained coatings was strongly dependent on the metal surface quality, impurities, wettability of the coating precursor solutions. Figure 6 shows the spinel coatings obtained on the porous metal supports and anode backbone. Corrosion testing of the half-cells with these coatings showed an insufficient protection against the oxidation of the FeCr metal particles, due to the poor adherence, discontinuous and non-uniform nature of the coatings. This indicates that the adherence and uniformity of these coatings required further optimization.

In summary, to exploit the potential of these coating materials, further development and optimization of the coating process is necessary so that continuous, uniform and adherent coatings that can offer the improved oxidation resistance to FeCr metal surface.

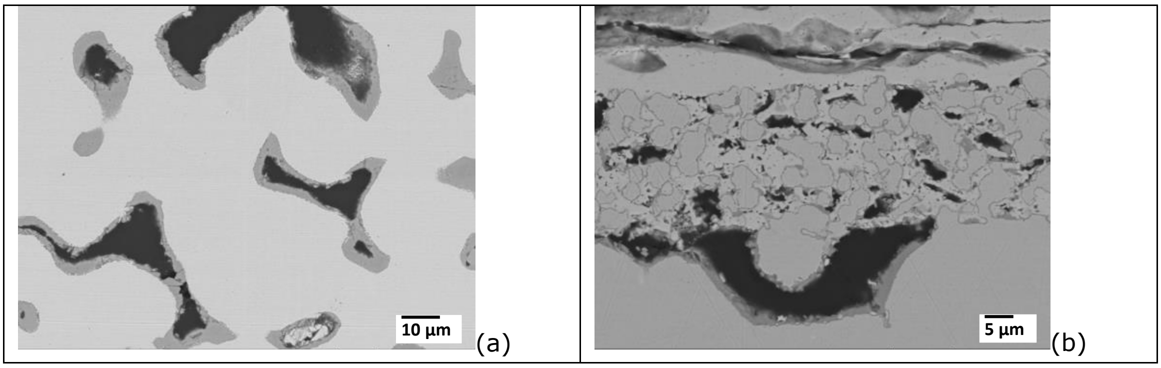

Figure 6: SEM micrograph of spinel coatings (a) metal support and (b) coating in the anode functional layer.

1.5.1.3 Development of alternative anode designs

In this third approach, the alternative anode designs based on electronically conducting ceramic materials such as doped $SrTiO_3$ are developed. These materials should be stable under highly reducing environment at high temperatures during cell processing, possess good electronic conductivity and compatible with other cell components. Among the doped $SrTiO_3$ materials, DTU focused on the development of Nb-doped $SrTiO_3$ (STN) materials and an EU partner, Univesity of St. Andrews (USTAN), focused on the (La,Sr)(Ti,Fe,Ni)O_3 (LSFNT) materials. The integration of the STN based anode designs is challenging due to the different sintering behaviour in cell processing conditions, resulting in poor adhesion with electrolyte and leaky electrolyte layer (Figure 7). Corrosion testing of the STN-FeCr anodes in MS-SOFC operating conditions indicated that the oxidation resistance of the FeCr in the anode designs is significantly improved. Introducing STN as a composite with the FeCr phase in the anode backbone led to a decrease in the formation of Cr_2O_3 on the FeCr particles. STN seem to block the inward diffusion of oxygen ions (if it covers the FeCr surface)[6]. However, Cr diffusion seems to occur at grain boundaries of the STN and allows for the formation of Cr-rich oxides in the pores of the anode and electrolyte.

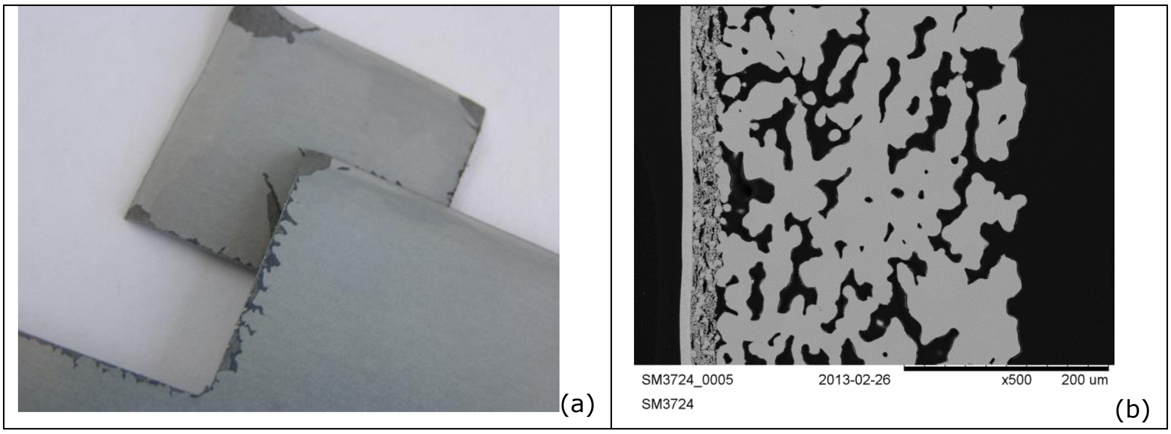

Figure 7: Metal supported half-cell with STN-FeCr anode backbone (a) after cutting the cell for microscopy and (b) SEM cross-section of the half-cell.

The electrochemical performance of the FeCr-STN based anodes showed the promise initially in terms performance and stability, particularly the later[6]. However, large scatter in the ASR of the cells (0.72 – 1.23 Ωcm^2) made the reproducibility quite challenging. The observed scatter is attributed to the variation in cell quality because of the challenges in integrating the STN material into the cells and non-uniform distribution of the Ni:GDC electrocatalyst material with in the anode backbone.

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[6] P. Blennow, B.R. Sudireddy, ÅH. Persson, T. Klemensø, J. Nielsen, K. Thydén, *Fuel Cells,* 13 (2013) 494.

The LSFNT anode material developed at USTAN showed the most promising characteristics required for their integration into MS-SOFCs. The special characteristic of these materials, exsolution of the nanoparticles, contributed to the improved adhesion with other cell components, and also probably enhanced their electrochemical performance. These materials possess good electrical transport properties and favourable sintering behaviour to be matched with other cell materials, making their integration into the MS-SOFCs successful. Different anode backbone designs are formulated using this material and their integration behavior was investigated to further develop the cells with novel anode backbones. These anode backbone designs consist of LSFNT, LSFNT-FeCr, LSFNT-FeCr-ScYSZ and LSFNT-ScYSZ. Among these designs, LSFNT-FeCr-ScYSZ anode backbone showed the most promising microstructure with significantly improved adhesion with electrolyte and metal support (Figure 8a). The addition of small amount of ScYSZ improved the adhesion between the anode backbone and the electrolyte significantly and the FeCr addition ensured adhesion with the metal support. Corrosion testing of the half-cells with FeCr-LSFNT based anode backbones in the MS-SOFC operating conditions showed negligible oxidation of the FeCr particles (Figure 8b). The LSFNT phase covered the FeCr metal particles, as in the case of FeCr-STN anode backbones, and prevented the oxidation of FeCr metal particles. The exact mechanism of protection needs to be establish through advanced analytical methods.

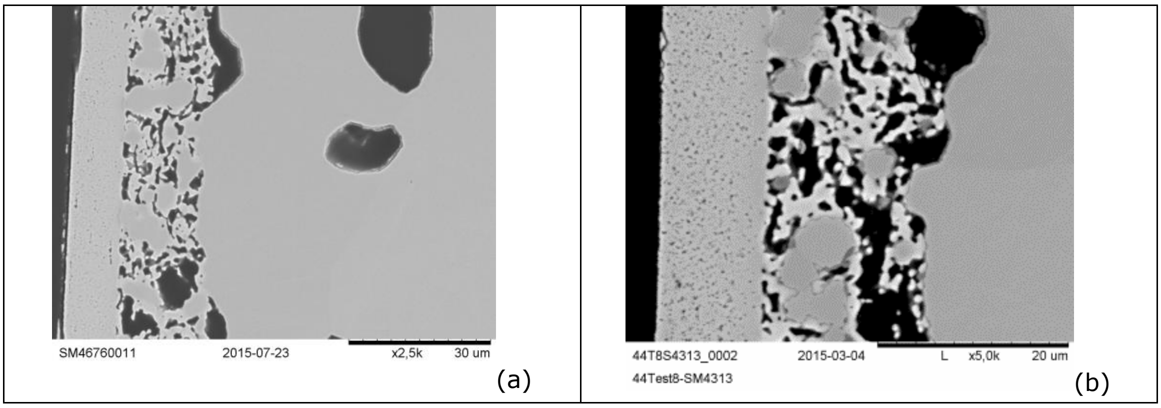

Figure 8: SEM micrograph of the half-cell (a) LSFNT-FeCr-ScYSZ anode backbone layer and (b) after corrosion testing at 650ºC for 250 h.

Electrochemical testing of FeCr-LSFNT based anode designs with Ni:GDC showed significantly improved performance and durability. The cells with most promising microstructure showed the performance of ASR equal to 0.57 Ωcm^2 with relatively good stability (Figure 9). The improved stability of these cells allowed the extraction of the electrochemical parameters for the modelling, which was a challenge with the MS-SOFCs using FeCr-YSZ anode backbones. Among the different titanate based anode designs, the LSFNT based anode designs showed superior performance and stability, paving the way for the development of robust MS-SOFCs.

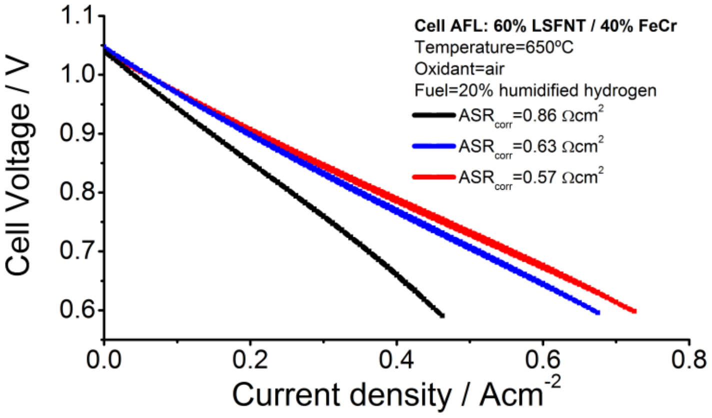

Figure 9: Performance characteristics of three different FeCr-LSFNT anode backbone based cells.

Excellent initial performance, an ASR of 0.32 Ωcm^2 at 650ºC, is obtained with FeCr-YSZ anode backbone, best among the reported performance for MS-SOFCs, when infiltrated with Ru:GDC electrocatalyst instead of Ni:GDC (Figure 10), however, the stability of these cells seems to be strongly dependent on the anode backbone microstructure and uniformity of the electrocatalyst coating[7].

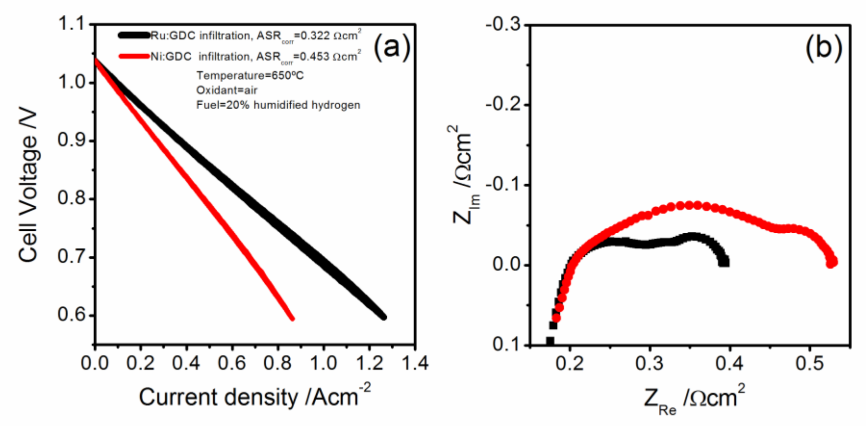

Figure 10: (a) Polarization curves and (b) Electrochemical impedance spectra of the Ru:GDC and the Ni:GDC infiltrated MS-SOFCs.

1.5.2 Development and integration of high performance and stable cathodes

The current cell design requires that the cathode is sintered in-situ during the initial start-up used for electrochemical testing. The in-situ "sintering" temperature is significantly lower than the temperature usually used for purely ceramic based intermediate and high temperature SOFCs. The different cathode materials, such as $(La_{0.6}Sr_{0.4})_{0.99}CoO_3$ (LSC), $(La_{0.6}Sr_{0.4})_{0.99}CoO_3{:}Ce_{0.9}Gd_{0.1}O_{1.95}$ (LSC:CGO), $La_{0.58}Sr_{0.4}Co_{0.2}Fe_{0.8}O_3$ (LSCF) and $La_{0.58}Sr_{0.4}Co_{0.2}Fe_{0.8}O_3{:}Ce_{0.9}Gd_{0.1}O_{1.95}$ (LSCF:CGO) are investigated and compared in the temperature range of 650 – 950 ºC. In contrast to LSCF, the LSC-based cathodes showed excellent sintering capabilities, electronic conductivity and performance. The polarization resistance (R_p) of the LSC based cathodes was 0.05 Ωcm^2, which to our knowledge is the best performance reported in the literature for a low temperature 800ºC in-situ sintered cathodes[8]. The challenge with this type of cathodes is to make them mechanically robust towards thermal cycling. Efforts were made to address this issue by decreasing the cathode and current collecting layer (CCL) thickness down to a thin ~15 µm LSC layer, acting both as a cathode and CCL. Testing of such thin LSC cathodes on single cells (16 cm^2 active area) showed promising and reproducible results with no observable delamination. The same cathode layer is integrated on to the larger cells (12 cm x 12 cm cells for TOFC stacks and 15.3 cm x 8.3 cm cells for EK stacks). The conditioning and initial performance testing of such stacks did not show any damage to the cathode, however, stability at thermal cycling is not tested due to lack of stacks that could be tested for long duration.

1.5.3 Component integration and cell manufacturing

The process for the integration of alternative cell components into the MS-SOFCs is optimized for different sizes of cells (12 cm x 12 cm and 15.3 cm x 8.3 cm) with good success. The challenges associated with upscaling the size of the cells are addressed successfully and good quality cells are fabricated. One of the major challenges encountered during the course of the project was the quality of the raw materials. Series of experiments using raw materials with different quality and distinguishable physical characteristics are carried out. The required sets of physical characteristics of the raw materials to fabricate the good quality cells are identified. Even though the whole process was time and resource consuming, it has pro-

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[7] B. R. Sudireddy et.al., *ECS Trans.,* 68 (2015) 1417

[8] J. Nielsen, P. Hjalmarsson, M. H. Hansen, P. Blennow, *J.Power Sources,* 245 (2014) 418

vided comprehensive understanding of the influence of quality, physical characteristics of raw materials on the cell quality.
Making the multilayered structures using tape casting process can be more cost-efficient if the layers can be cast simultaneously, particularly for mass production. During the course of the project, an environmentally friendly co-casting process, where all the layers were casted simultaneously, is developed with different variations (wet-on-dry, semi wet-on-wet and wet-on wet with wet-on-dry and semi wet-on-wet being most successful).

Despite the significant challenges encountered in the availability of sintering furnace, the lack of suitable raw materials quality and the change in cell design by the new stack building and development partner, every effort is made to minimize their impact on delays in the cell deliveries to the partners. Whenever suitable, the environmentally friendly and mass producing co-casting process is used for the manufacturing of cells and components. A summary of the cells and components made in different sizes for various activities are presented in Table 1.

Table 1: Summary of cells fabricated and supplied to different partners during the project.

Cell type	**Cell size (cm)**	**No. of cells**	**WP/Partner**	**Remarks**
G1	12 x 12	234	WP5/TOFC	To evaluate the cells in stacks and to optimize the sealing process
G1/G2	12 x 12	120/15	WP2/DTU	To be delivered to TOFC (WP5), TOFC closed
G1	14.5 x 29	1	WP2/DTU	For demonstration
G1/G2/G3	4 x 5	27/12/12	WP3/KIT	To test electrochemical performance of cells
G1/G2/G3	5 x 5	17/4/4	WP3/AVL	To optimize the THDA monitoring model
G1	7.6 x 4.15	15	WP5/EK	To optimize the sealing process
G1	7.6 x 8.3	10	WP5/EK	To optimize the sealing process
G1/G3	15.3 x 8.3	31/8	WP5/EK	To evaluate the cells in new stack design

The demonstration of the mass volume production (more than 500 cells of different sizes), flexibility with cell geometry (sizes ranging from 5 cm x 5 cm – 14.5 cm x 29 cm) and reasonably high yield of the cells indicates that the current cell fabrication technology is upscalable and industrially relevant. The co-casting and co-sintering of different layers in the cells makes the cell fabrication process significantly cost-efficient and economic.

1.5.4 Mechanical testing of metal supported cells

The mechanical testing of the metal supported cells payed particular attention to elastic response of the metal support and electrolyte as a function of temperature, creep behavior of metal supports and interaction with oxidation and 3D simulation of the creep in metal supports. The elastic modulus at room temperature before the thermal treatment is measured to be 60.0±0.3 GPa. The Young's modulus "E" decreased more or less linearly with an increase in the temperature down to 32.5±0.6 GPa at 700 °C. Thermal treatments slightly increased the E due to the formation of oxide scale. The elastic moduli of the ScYSZ electrolyte is found to have a significant peak around 450 °C, where the stiffness is believed to be lowered due to atomistic reorganizations. This will influence the stress distribution amongst the layers over a temperature cycle, but as it occurs mostly the purely elastic temperature range, little impact is expected. A stack model is however needed to fully evaluate the impact.
For the metallic components of the MS-SOFC stack, i.e. the interconnect (IC) steel and the metallic support (MS), the creep properties were measured and parameters for the constitutive creep laws for both primary and secondary creep were extracted from the measure-

ments. The secondary creep is important for long-term steady operation, where as the primary is highly important for load cycling. In the non-oxidized state, both IC and MS samples showed same dependency with stress (Figure 11). In the oxidized state the material behavior became complex. The oxide scale formed from corrosion is a less creeping ceramic coating and thus, an oxide scale minimizes the creep rate. However, if higher stresses are applied, the oxide scale peals off, and the creep rate increases compared to the non-oxidized material. This mechanism also increases the corrosion rate, as the protection from the oxide scale is lost and thus results in further corrosion[9]. Infiltration minimizes the corrosion and thus also the amount of corrosive scale. Therefore, the infiltrated metal support creeps faster. Aging treatment of the infiltrated samples, found to be decreasing the creep rate.

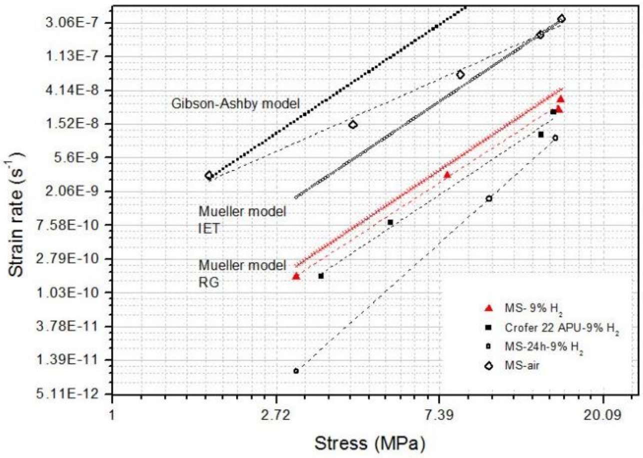

Figure 11: Norton plots (secondary creep rate vs. stress) of porous ferritic steel supports and Crofer. Fitting models are also reported.

1.5.5 Stack design, stack building and testing

The focus of the stack-related work at TOFC was on the development of improved sealing technology and the implementation of a new stack design for metal supported cells. TOFC showed successful integration of metal-supported cells having high power density at lower temperatures and operating for a few hundred hours into the stacks. However, the efforts to test these stacks revealed that successful stack operation could not be guaranteed for every stack because of failure modes that are initiated during the very first treatment of the stack at elevated temperature. Therefore, in addition to project goals of high performance and larger stack sizes, the principle objective of improving stack treatment without damage was brought into focus. During the project, TOFC made changes in stack sealing strategy, geometrical and material designs, as well as procedures for high temperature treatment.

1.5.5.1 Development of stack design

Along the stack components materials data, the stack design development utilized computer simulation tools to build a multiscale model of metal supported stacks at TOFC. Specialized FEM and other mechanical simulations have been investigated in order to understand the creep behavior of the stack interconnects. The results yielded important information about the sensitivity of cell mechanics to creep phenomena at different loads and temperatures. Preliminary conclusions / findings from this work:

- Modelling of metal support creep based on Norton Power law constitutive formulation seem untrustworthy – improved equations are needed.
- More complex flow rule formulations should be evaluated, which take into account different dislocation and diffusion mechanisms in different temperature regimes.
- Effect of coatings and impregnations are not sufficiently understood in relation to composite creep behaviour description

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[9] D. Boccaccini et.al., *Int. J. Hydrog. Energy*, 39 (2014) 21569

Together with mechanical properties measurement, various mathematical modelling tools have been developed for design optimization. Most notable among these is a flow-homogenization tool (see Figure12), which is a universally applicable tool with the intention to be used to optimize flow patterns.[10] This was especially important as new interconnect geometries were to be considered, since new geometries can contain complex patterns—and yet require fast feedback concerning stack operation in order to apply appropriate design tweaks. These tools are still used at Haldor Topsøe A/S (HTAS), a parent company of TOFC for SOEC stack design development.

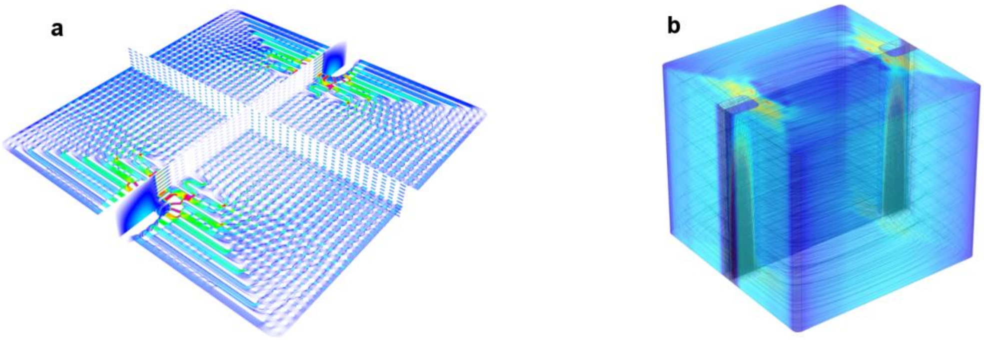

Figure 12: Homogenization process of fluid flow in a stack. a) Detailed Navier-Stokes model of a 10-unit-cell stack. b) Homogenized model of a 75-unit-cell stack based on simplified Darcy flow in porous media. The homogenized model was obtained from the full Navier-Stokes solution using topology optimization procedure.

Simultaneously an EU partner, AVL, continued the development of another tool called AVL FIRE ® v2014, which contains a comprehensive 3D electrochemical model for the simulation of solid oxide fuel cells. The model is applicable to single cells, stacks, steady and transient analyses with pure hydrogen or hydrocarbon as fuel. In future, the model shall be extended in order to account for degradation mechanisms (e.g. carbon formation) and in order to enable stress and deformation analysis.

1.5.5.2 Development and building of stacks

The focus of the stack development work was to develop 10-25 cell stacks with optimal compression forces, improved laser welds (of cells to interconnects), improved stack conditioning and glass sealing procedures, capability of internal reforming in the stack, and to eventually test them for at least 1000h. The work resulted in improvements on all aspects of the stack fabrication steps and conditioning, which lead the development of promising stacks that are intended for stack testing towards the end of 2014. However, due to the closing of TOFC in August 2014 the stacks were unfortunately never tested. Some of the development highlights are given below:

Several stack development trials are conducted during the project, with variations in stack composition and treatments. The stack treatments included variations in the application and duration of compression forces, in order to determine the importance of mechanical force and metal creep. The conclusion from those tests and others is that too little load adversely affected stack start-up, whereas higher loads could be tolerated in some cases without catastrophic failure. Therefore, further efforts are focused to find an optimal compression force, at which there is no cross-over leakage and also no severe creep. This is meant to develop and implement a new stack design with the purpose of having minimal impact of creep, so that safer (higher) compression forces will induce only a low amount of creep.

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[10] Elesin Y., Madsen, M. F., Petersen T. K., Topology Optimization based homogenization technique for stack designs with complex geometry. In: Christiansen N, Hansen JB, editors. Proc. 11th Eur. SOFC SOEC Forum 2014, European Fuel Cell Forum AG; 2014, p. B11.15–B11.23.

Laser welding of cells to interconnects (IC): The laser welding parameters for welding the metal supported cells to ICs are significantly improved. With the introduction of a "double-weld" together with improved laser weld parameters the leak rates from the cell-IC component became sufficiently low for proper implementation into stacks.

Stack birth and conditioning: This step involved assembly of the cell-IC single-repeat units into stacks and forming a good seal between the cell of one cell-IC component and the IC of another cell-IC component (see Figure 13). Here, progress is made in finding a proper procedure including choosing an appropriate glass material, finding a suitable temperature profile during the sealing process, as well as optimal compression during the sealing step to form a leak tight stack without damaging the electrolyte.

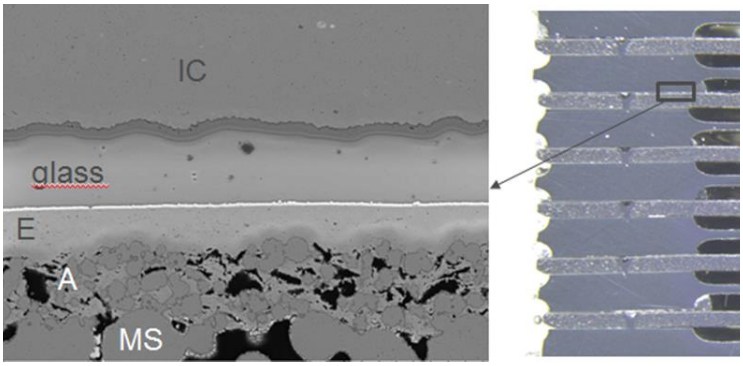

Figure 13: Cross-section of a trial 6-cell stack at the edge (right image) with the corresponding good glass seal (left image). Note the good adhesion of the glass to both the IC and the electrolyte (E).

Cathode sintering - ex-situ vs. in-situ: One of the challenges with the MS-SOFC technology developed in the project was how to solve the issue with sintering of the cathode. One of the suggested changes included a new strategy of *ex-situ* cathode sintering, which required several modifications in the stack fabrication procedure. A patent application was completed and submitted on this strategy. TOFC is eventually able to prove that an *ex-situ* process indeed is possible, however, the process is unfortunately deemed to be too difficult to use with respect to ensuring sufficient reproducible quality of the cathode after stack assembly, as well as from a cost perspective when the process is going to be up-scaled.

An alternative route with *in-situ* cathode sintering, where the cathode is sintered when the stack has been assembled and is sealed with glass at elevated temperatures, was therefore developed. The *in-situ* sintering route required testing and implementation of new glass materials for sealing and new methods for their application and processing (heat treatment and compression). New insights into the different fabrication steps of the stacks made it eventually possible to fabricate a very promising stack, which after assembly showed promising low leak rates (see Figure 14). The developed stack fabrication route resulted in a stack that could be handled without compression and was approved with regards to the leak rates. The stack was unfortunately never characterized in a stack test due to the closing of TOFC at the same time as this stack had been produced.

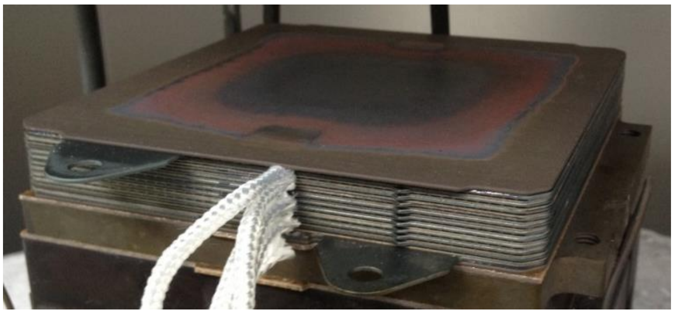

Figure 14: A 10-cell stack with MS-SOFC after assembly and conditioning with in-situ sintering of the cathode. The stack was approved with regards to the leak rates and was intended for stack characterization as the next step.

After the closure of TOFC, an EU partner ElringKlinger A/G (EK), Germany took over the tasks of stack development and testing in the project. EK has successfully integrated the metal supported cells developed in METSAPP project into their stack design. Test of the stacks could not accomplished due to shortage of time in EU project. However, successful integration of the metal supported cells into two different stack designs by two different partners (TOFC and EK) demonstrates that the metal supported cells are adaptable for different stack designs.

1.5.6 Answer to the problem statement

The main objective of the METSAPP was to develop the novel cells and stacks based on a robust and reliable up-scalable metal supported technology. A robust cell design with an ASR_{cell} of 0.32 Ωcm^2 at 650°C is demonstrated, which is well below the project target of 0.5 Ωcm^2. To our knowledge, this is the lowest ASR reported for planar MS-SOFCs. The demonstration of the large area cells (> 300 cm^2) fabrication is accomplished through the fabrication of cell with an area larger than 400 cm^2. The scalability, adaptability and cost-efficiency of the cell fabrication process is demonstrated through the fabrication of more than 500 cells with different sizes using an environmentally friendly co-casting process using few processing steps. Successful integration of the cells into two different stack designs indicates the adaptability of the cells and modularity of the stacks. The target set out for the stability of the cells is considered quite ambitious and addressing this target requires further optimization of the cells. Testing of the stacks and verification of the robustness and stability of the stacks could not be accomplished, as unforeseen challenges faced in the project caused delayed the building of stacks.

The overall goal of developing the robust MS-SOFCs is addressed with promising results through the development of novel anode and cathode designs that are high performing with relatively good stability. Furthermore, the successful integration of the cells into two different stacks indicates the modularity and mechanical robustness of the stacks.

1.5.7 Expected impact and final assessment

In the Table 2, the proposed expected outcome of the METSAPP project and assessment of the achieved outcome is presented.

Table 2: Expected outcome from the project and final assessment.

Parameter	**Expected outcome**	**Assessment**
Cell design	Alternative cell design to the SoA electrode and/or electrolyte supported design.	An alternative cell design based on metal support with novel electrode architecture demonstrated.
Cost efficiency	Reduce the cost of the SOFC compared to Ni-YSZ based cells	A cheaper FeCr alloy based supports are successfully integrated instead of expensive Ni-YSZ supports. The optimized cell fabrication process consists single sintering step, which reduces the cell fabrication cost significantly.
Scalability	Scalable cell manufacturing process	An environmentally friendly co-casting process is developed and scalability is demonstrated through the fabrication of more than 500 cells
Large cell area	> 300 cm^2 cell area	> 400 cm^2 cell area demonstrated through the fabrication of cell with 14.5 cm x 29 cm dimensions
Thermal cycling	Robustness of the cells and stacks towards thermal cycling	No physical damage to the cells and/or cell components observed on cell level. The testing requires on stack level.
Tolerance towards temperature gradients and operation cycles	Tolerance towards temperature gradients and operation cycles	The cells used in single cell testing (5 cm x 5 cm) are not large enough to assess this. Conditioning tests on stacks did not show any damage to stacks and/or stack components. Long-term tests under current are required to assess this parameter.

1.5.8 Environmental impact

The MS-SOFC technology facilitates the operation of SOFC at much lower temperatures compared to the conventional anode supported cells, which would facilitate the use of cheaper and inexpensive materials. Apart from the lower CO_2 emission advantages from the lower temperature operation, the inherent advantage of SOFCs will further reduce the CO_2 emissions. The other inherent advantages of SOFCs include, absence of NO_x and SO_2 emissions along with easier/cheaper capture and processing of CO_2 due its availability in the concentrated from during the SOFC operation.

The other environmental aspect addressed in the project is the replacement of expensive and toxic Ni usage in the MS-SOFCs. Both the anode backbone and cell support are fabricated using the environmentally friendly FeCr ferritic alloys with the addition of YSZ and/or titanate ceramics, also considered environmentally benign, in the anode backbone, thus the NiO is removed from the cell components. Furthermore, during the process of developing the scalable cell manufacturing process, the toxic organic additives in the tape casting slurries, such as phthalates, are exchanged with environmentally friendly additives making the cell fabrication process friendlier. The development such process with non-toxic additives for the MS-SOFC component fabrication can be transferred to the components fabrication in any other technologies that uses tape casting for component fabrication, thus results in much broader impact beyond this project.

1.5.9 Dissemination of project results

The work carried out in this project has resulted in 9 peer reviewed articles, 7 conference proceedings, 2 patents. In addition to this, project results have been presented at several international conferences and workshops inform of oral or poster presentations. The details of the dissemination are listed below:

Publications

1. P. Blennow, J. Hjelm, T. Klemensø, S. Ramousse, A. Kromp, A. Leonide and A. Weber, *J. Power Sources,* **196** (2011) 7117.
2. B. J. McKenna, N. Christiansen, R. Schauperl, P. Prenninger, J. Nielsen, P. Blennow, T. Klemensoe, S. Ramousse, A. Kromp and A. Weber, *Fuel Cells,* **13** (2013) 592.
3. P. Blennow, B.R. Sudireddy, Å. H. Persson, T. Klemensø, J. Nielsen and K. Thydén, *Fuel Cells,* **13** (2013) 494.
4. A. Kromp, J. Nielsen, P. Blennow, T. Klemensø and A. Weber, *Fuel Cells,* **13** (2013) 598.
5. J. Nielsen, P. Hjalmarsson, M. H. Hansen, P. Blennow, *J. Power Sources,* **245** (2014) 418.
6. D. N. Boccaccini, H. L. Frandsen, P. Blennow, B. R. Sudireddy, Å. H. Persson, K. Kwok, P. Vang Hendriksen, *Int. J. Hydrog. Energy,* **39** (2014) 21569.
7. G. Reiss, H.L. Frandsen; W. Brandstätter; A. Weber, *J. Power Sources,* **273** (2014) 1006.
8. G. Reiss, H.L. Frandsen; A. H. Persson; C. Weiß; W. Brandstätter, *J. Power Sources,* **297C** (2015) 388.
9. K. Kwok, D. Boccaccini, Å. H. Persson, H. L. Frandsen, *Int. J. Solids and Structures,* **78-79** (2016) 38.

Conference Proceedings

1. P. Blennow, Å. H. Persson, J. Nielsen, B. R. Sudireddy, T. Klemensø, *Proceedings of 10th European SOFC Forum,* **Ch.7** (2012) 72.
2. N. Christiansen, S. Primdahl, M. Wandel, S. Ramousse, A. Hagen, *Proceedings of 10th European SOFC Forum,* **Ch.3** (2012) 11.
3. P. Blennow, B. R. Sudireddy, Å. H. Persson, J. Nielsen, R. Sachitanand, J. Froitzheim, *ECS Transactions,* **57** (2013) 771.
4. N. Christiansen, S. Primdahl, M. Wandel, S. Ramousse, A. Hagen, *ECS Transactions,* **57** (2013) 43.
5. D. Boccaccini, P. Blennow, H. L. Frandsen, P.V. Hendriksen, *Proceedings of Fifth European Fuel Cell Technology & Applications Conference (EFC 2013),* (2013) 291.
6. B. R. Sudireddy, J. Nielsen, K. T. S. Thydén, Å. H. Persson, K. Brodersen, *ECS Transactions,* **68** (2015) 1417.
7. J. Nielsen, B. R. Sudireddy, A. Hagen, Å. H. Persson, *ECS Transactions,* **68** (2015) 1361.

Patents

1. Brandon J. McKenna, Cliver Klitholm, Air electrode sintering of temporarily sealed metal-supported solid oxide cells, EP2830127, 2013.
2. Brandon J. McKenna, Rainer Küngas, Tobias Holt, Peter Blennow, Metal-supported solid oxide cell, EP2808932, 2013.

Conference/workshop presentations

1. P. Blennow, Å. H. Persson, J. Nielsen, B. R. Sudireddy, T. Klemensø, Infiltrated SrTiO3:FeCr-based anodes for metal-supported SOFC, 10th EFCF, 26-29 June, 2012, Lucerne, Switzerland.
2. N. Christiansen, S. Primdahl, M. Wandel, S. Ramousse, A. Hagen, Status of the SOFC development at TOFC and DTU, 10th EFCF, 26-29 June, 2012, Lucerne, Switzerland.
3. P. Blennow, Metal-supported Solid Oxide Fuel Cells: Next generation SOFC?, Lund Fuel Cell Research Network, 30 March, 2012, Lund, Sweden.
4. P. Blennow, C. Graves, B. R. Sudireddy, M. Mogensen, New materials for solid oxide cells - nanostructured electrocatalysts by infiltration and/or exsolution, SERC bi-annual meeting, 11 November, 2012, Roskilde, Denmark.
5. Å. H. Persson, P. Blennow, T. Stegk, T. Klemensø, Fabrication and tailoring of porous metal membranes by tape casting, Nordic conference on ceramics and glass technology, 6-7 December, 2012, Roskilde, Denmark.
6. P. Blennow, B. R. Sudireddy, Å. H. Persson, J. Nielsen, R. Sachitanand, J. Froitzheim, Investigation of Cu-based infiltration coatings for metal-supported SOFC, SOFC-XIII, 6-11 October, 2013, Okinawa, Japan.
7. N. Christiansen, S. Primdahl, M. Wandel, S. Ramousse, A. Hagen, Status of SOFC development at TOFC and DTU, SOFC-XIII, 6-11 October, 2013, Okinawa, Japan.
8. D. Boccaccini, P. Blennow, H. L. Frandsen, P.V. Hendriksen, Creep behaviour of porous high Cr ferritic steel solid oxide fuel cell metal supports, EFC 2013, 11-13 December, 2013, Rome, Italy.
9. N. Christiansen, Topsoe Fuel Cells Development, 9th Annual International Hydrogen & Fuel Cell Conference, 20-21 March, 2013, Birmingham, UK.
10. N. Christiansen, Progress and Challenges of Metal Supported SOFC, ICACC 14, 26-31 January, 2014, Daytona Beach, FL, USA.
11. N. Christiansen, Solid Oxide Fuel Cells Challenge in the development from science to Technology and Industrialization, Fuel Cells 2014 Science & Technology Conference – A Grove Fuel Cell Event, 3-4 April, 2014, Amsterdam, The Netherlands.
12. N. Christiansen, New Generation Solid Oxide Fuel Cells, CIMTEC 2014, 15-19 June, 2014, Montecatini Terme, Italy.
13. B.R. Sudireddy, S. Veltzé, J. Nielsen, P. S. Jørgensen, T. Ramos, Nb-doped $SrTiO_3$ and Gd-doped CeO_2 composites - potential anodes for solid oxide fuel cell applications, 11th CMCEE, 14-19 June, 2015, Vancouver, Canada.
14. B. R. Sudireddy, J. Nielsen, K. T. S. Thydén, Å. H. Persson, K. Brodersen, Investigation of novel electrocatalysts for metal supported solid oxide fuel cells - Ru:GDC, SOFC-XIV, 26-31 July, 2013, Glasgow, UK.
15. J. Nielsen, B. R. Sudireddy, A. Hagen, Å. H. Persson, Performance Factors and Sulfur Tolerance of Metal Supported Solid Oxide Fuel Cells with Nanostructured Ni:GDC Infiltrated Anodes, SOFC-XIV, 26-31 July, 2013, Glasgow, UK.

Apart from the above-mentioned forms of dissemination, the project is also presented in a workshop organized by FCH JU RAMSES project in France on 22 May 2014, with other FCH JU project participants that are working on metal supported cells. Nationally, the project summary is also sent for publication in magazine called "FiB" that publishes the progress of the Danish national projects in energy sector.

1.6 Utilization of project results

The development targets necessary for the commercialization of the SOFC technology are presented in the Danish roadmap and strategy 2013-25.[11] The progress achieved in the present project forms an important step towards achieving these SOFC development targets. The advancements made in the development of robust, stable and high performance electrode materials forms the basis for developing the robust MS-SOFCs for both stationary and mobile applications. The progress made in the development of MS-SOFC stack designs, stack

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[11] Strategi for udvikling af SOFC-brændselsceller 2013-2025, Partnerskabet for Brint og Brændselsceller, http://www.hydrogennet.dk/sofc-strategi1/

building, and the adaptability demonstrated in the integration of the cells into different stack designs can be utilized by TOFC (HTAS) and ElringKlinger A/G for further advancements of this technology towards the market. It is already showed that the mathematical tools developed in the project for MS-SOFC stack design optimization can also be used for the stack development for SOECs.

The results obtained in the project are published in the refereed journals, presented in international conferences and workshops, thus the reported knowledge will benefit the wider scientific community in further advancing the MS-SOFC technology towards the commercialization. Furthermore, two patents are obtained based on the knowledge obtained from the results of the project.

1.7 Project conclusion and perspective

The development of robust metal supported fuel cells and stacks is addressed in the project. The key achievements are summarized in the following:

- The critical parabolic rate constants for oxidation of FeCr alloys is established and can be used as a guideline for alternative alloys development for enhanced lifetime.
- The nanostructured oxidation protective coatings developed for FeCr-YSZ anode backbones did not affect their functionality negatively. Chromite and spinel based coatings showed promise and further optimization of these coatings was necessary to improve their efficiency.
- Alternative anode materials based on doped strontium titanate are developed and anode backbones based on LSFNT are successfully integrated into MS-SOFCs.
- Doped strontium titanate anodes showed significantly improved oxidation resistance of the FeCr metal particles under MS-SOFC operating conditions.
- LSFNT based anode designs with Ni:GDC electrocatalyst showed superior electrochemical performance (ASR: 0.57 Ωcm^2 and stability.
- MS-SOFCs with FeCr-YSZ anode backbones and Ru:GDC electrocatalyst showed the best electrochemical performance (ASR:0.32 Ωcm^2) for planar cells. The degradation of these cells require further improvement.
- High performance (polarization resistance: 0.05 Ωcm^2) and stable cathode designs, based on LSC, are developed and integrated.
- Integration of the novel cell components on various cell sizes (from 25 cm^2 to more than 400 cm^2) is successfully demonstrated.
- Scalable metal supported cell fabrication process for mass production (more 500 cells in different sizes are fabricated) is successfully demonstrated through the development of environmentally friendly and cost-efficient co-casting process.
- Adaptability of cell fabrication process for various sizes (from 25 cm^2 to more than 400 cm^2) is successfully demonstrated.
- Different mathematical tools for MS-SOFC stack design are developed and used for stack development.
- Metal supported cells are successfully integrated into two different stack designs (TOFC and EK) and stacks are built, showing the adaptability of the cells for different stack designs.
- The process for sealing the stacks and stack conditioning is developed with optimized parameters resulting in good quality stacks.
- Methods and parameters for both *ex-situ* and *in-situ* sintering of the cathodes in stacks are developed.

Significant insight is gained into the various options to improve the robustness of the anode designs during the project. Some options, such alternative anode materials, proved to be more viable than others. Protective coatings for FeCr based anode designs showed promise, however, requires improvement in the quality of the coatings to realize their full efficiency against oxidation. Anode designs based on doped strontium titanate materials proved to be most promising in terms of oxidation resistance with good electrochemical performance. The excellent initial performance of the novel anode/cathode designs can facilitate the commercialization of portable MS-SOFCs for the *niche* applications that require short-life time (ca. 5000 h). The realization of long-term applications for the MS-SOFCs require further development to reduce their degradation rates through the stable component development in future projects. The scalability, adaptability, cost-efficiency of the cell fabrication process and the adaptability of the metal supported cells for different stack designs are expected to encourage and attract the attention of the SOFC market players. Extending the electrochemical performance observed on cell level to the stack level and establishing the MS-SOFC stack technology would be a natural next-step towards the commercialization of the MS-SOFC technology.

Annex

1. Appendix P: the financial statement has been updated on the www.forskel.dk and is attached along with this report.
2. The project website has been created for FCH JU METSAPP project and the details can be seen at the project website: www.metsapp.eu