

Energimyndighetens titel på projektet – svenska Kostnadseffektiva IT-SOFC bränsleceller för mobila applikationer	
Energimyndighetens titel på projektet – engelska Cost effective IT-SOFC for mobile applications	
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Nyckelord: 5-7 st SOFC, Fast oxid bränsleceller, Bipolära plattor, Fordons applikationer	

Förord

The project was founded by The Swedish Energy Agency, AB Sandvik Materials Technology, Elcogen Oy and Volvo Technology AB

Innehållsförteckning

Sammanfattning	1
Summary	2
Inledning/Bakgrund	3
Genomförande	3
Resultat	4
Diskussion.....	15
Publikationslista.....	16
Bilagor	18

Sammanfattning

Fastoxidbränsleceller (SOFC) genererar ren ström med hög verkningsgrad. Teknologin har stor potential som APU-enheter (Auxiliary Power Units) i lastbilar eller som ”range extenders” i elbilar. Idag begränsar livslängde och det höga priset per kW storskalig SOFC-implementering. Försämring av SOFC-stacken och en betydande del av de totala kostnaderna är kopplade till de bipolära plattorna.

Fokus för detta projekt var att utveckla högpresterande kostnadseffektiva bipolära plattor, vilket möjliggör förbättrad SOFC-prestanda och samtidigt en avsevärd sänkning av priset.

Projektet resulterade i betydande framsteg i utvecklingen av en kostnadseffektiv lösning för de bipolära plattorna i SOFC. Sandviks världsunika förbelagda Co/Ce-belagda material integrerades framgångsrikt i en stack från Elcogen och kördes i 3000 timmar. Nedbrytningshastigheten är i samma storleksordning som Elcogens ”state of the art” lösning. Detta är ett stort genombrott och ett ”proof of concept” för användningen av förbelagt stål i SOFC-stackarna. Det förbelagda stålkonceptet möjliggör produktion av material i hög volym på ett mycket kostnadseffektivt sätt. Resultaten som erhålls i detta projekt kommer att underlätta kommersialiseringen av SOFC-teknologi till en bredare marknad genom att öka livslängden och minska kostnaderna för SOFC-stackar.

Konsortiet i detta projekt omfattar hela värdekedjan, från komponent- och SOFC-tillverkare (Sandvik Materials Technology och Elcogen) till slutanvändare (Volvo). Chalmers är projektkoordinator och ansvarig för både laboratoriestudier och materialkaraktärisering. Projektperioden var 2018-03-06 till 2022-05-31. Den totala budgeten var 9,5 MSEK.

Summary

Solid Oxide Fuel Cells (SOFCs) generate clean electricity with high efficiency. This technology has huge potential as Auxiliary Power Units in Trucks or as EV range extender. Today, the high price is limiting large-scale SOFC implementation. Degradation of the SOFC stack and a substantial part of the total costs are linked to the interconnect material.

The focus of this project was to develop high performing low-cost interconnects, allowing for improved SOFC performance and at the same time a substantial reduction in price.

The project resulted in significant progress in developing a low-cost interconnect solution for SOFC. Sandvik’s world unique pre coated Co/Ce coated material was successfully integrated into a stack from Elcogen and operated for 3000h. The degradation rate is in the same range as the Elcogens state of the art (SoA) solution. This is major breakthrough and a proof of concept for the use of pre-coated steel in the SOFC stacks. The pre coated steel concept allows for high volume production material in a very cost-effective way. The results obtained in this project will facilitate the commercialization of SOFC technology to a broader market by increasing the lifetime and decreasing the cost of SOFC stacks.

The consortium of this project includes the complete value chain, from component and SOFC manufacturers (Sandvik Materials Technology and Elcogen) to end user (Volvo). Chalmers is the project coordinator and responsible for both laboratory

studies and material characterization. The project period was 2018-03-06 to 2022-05-31. The total budget was 9,5 MSEK.

Inledning/Bakgrund

Solid oxide fuel cells (SOFC) are attractive power units for mobile applications, like auxiliary power units or range extenders, due to high electrical efficiencies and fuel flexibility. SOFC have in contrast to PEM fuels cells an all solid electrolyte which requires a much higher operating temperature but also results in a greater fuel flexibility. The drawback of higher temperature are several thermally activated degradation processes as well as need for startup heating and insulation. The technology used in this project focuses on intermediate temperatures (600- 700°C) which makes IT-SOFC much more suitable for mobile applications.

The main challenges for implementation of the SOFC technology in mobile applications are the cost and life time of the stack. Degradation of the SOFC stack and a substantial part of the total costs are linked to the interconnect material.

This project has focused on more durable and economic interconnect materials employing low-cost substrates and coatings. With the focus on developing robust highly efficient Solid Oxide Fuel Cells (SOFCs) at a substantially lower price, this environmentally friendly technology can be implemented in a much larger scale in the future.

What is unique with this consortium is that the two material manufacturers (Sandvik and Elcogen) have the capacity to start mass production from day one, making this project highly relevant. This project contributes to several of the targets in the program, defined within “road map FFI”. Fuel cells in general can achieve very high electrical efficiency and the technology is a attractive candidate for both improvement of the efficiency, and reduction of emissions from the transport sector. In contrast to for example a combustion engine, fuel cells generates electricity through an electrochemical process, hence no moving parts (noiseless) and no formation of NO_x, SO_x, or particulate matter emissions (emission reduction). A specific advantage of SOFC compared to other fuel cell types is that SOFC are not limited to hydrogen fuel, but can also operate on hydrocarbon fuels, including a variety of renewable fuels.

Genomförande

This project consisted of six work packages (WP) that combined enable the development of coated interconnects for highly efficient cost-effective Solid Oxide Fuel Cells designed for APU applications. In WP1 the most important parameters were selected in order to lower costs. In WP2 coated interconnects were produced based on the analyses from WP1. In WP3 lab tests on a component level were carried out and in WP4 these materials were analysed. In WP5 the most promising

interconnect materials was selected for real stacks test followed by post-mortem analyses of these stacks in WP6.

Feedback loops between these six work-packages allowed for several iterations during the period of the project, with the final aim to have developed substantially cheaper Solid Oxide Fuel Cells for mobile applications at the end.

The project consisted of four phases. Phase 1 is a cost analysis to define target costs as well as a definition of crucial parameters in order to mass produce low-cost interconnects. In phase 2, the concept of pre-coating will be evaluated. In phase 2, stack tests with pre-coated Crofer 22 APU were conducted, and post mortem-analysis of both pre- and post-coated stacks was carried out as a comparison. In phase 3, coatings custom-made for low-cost steels was developed, focus was on the low cost steel AISI 441. The coatings evaluated included Cu/Ce and Cu/Mn/Ce as well as including Co/Ce and Elcogens state of the art coating. The coatings were tested both in labscale and in stack tests. After each stack test, post-mortem analyses were carried out.

The consortium of this project includes the complete value chain, from component and SOFC manufacturers (Sandvik Materials Technology and Elcogen) to end user (Volvo). Chalmers is the project coordinator and responsible for both laboratory studies and material characterisation.

Resultat

Novel coatings

For the SOFC technology to be successful, two main challenges must be overcome: a) the long-term stability of the system; and b) the material cost. The choice of interconnect material strongly influences these factors. Cr(VI) evaporation from the steels and the use of cheap stainless steels, such as AISI 441, may lead to shorter material life-spans but are more cost-effective and they undergo severe corrosion when exposed to high temperatures and humid atmospheres. One strategy to overcome these problems is the use of coatings. Among the state-of-the-art coatings is a Ce/Co coating. Copper-based coatings are investigated to decrease further the cost. Copper is seven times cheaper than cobalt (10 USD/kg for Cu vs. 70 USD/kg for Co).

In the project we have investigated a range of different coatings and two deposition techniques. The conventional Thermal spray coating and the much more cost efficient novel on-line PVD technique developed by Sandvik. Physical Vapour Deposition (PVD)-coated samples with Ce/Co, Ce/Cu, and Ce/MnCu, and Thermal Spray (TS)-coated Mn/Co, Cu and Mn/Cu and AISI 441 steel samples were exposed at 650°C for up to 1,000 h in the laboratory. The chromium evaporation, the oxidation rate and the Area Specific Resistance were measured.

Compared to uncoated samples, coated samples showed a much lower level of Cr(VI) evaporation throughout the exposure (see Figure 1). On average, PVD Ce/Cu-coated and Ce/MnCu-coated samples exhibited equivalent levels of Cr(VI) evaporation. In contrast, the state-of-the-art Ce/Co coating displayed a slightly lower Cr(VI) evaporation level. This difference should not be over-interpreted, as all of the values are extremely low.

TS Cu-coated samples, TS Mn/Cu-coated samples, and TS MCO-coated samples displayed similar Cr(VI) evaporation rates after 1,000 h of exposure to humid air, compared to the PVD coatings. The TS coatings appear to exhibit lower Cr(VI) evaporation level than PVD coatings. However, in this comparison one has to take into consideration that the TS coatings have coated edges while the PVD coated samples have uncoated edges.

To allow for a direct comparison PVD Ce/Co-coated samples with coated edges were exposed to the same conditions. After 1,000 h of exposure at 650°C in humid air, the PVD Ce/Co-coated samples with coated edges displayed a similar Cr(VI) evaporation rate as the corresponding TS coatings. PVD coatings seem to be as efficient as TS coatings when the edges are coated, while the coating thickness is 10-fold thinner for PVD coatings ($\sim 1 \mu\text{m}$ for PVD compared to $\sim 10 \mu\text{m}$ for TS).

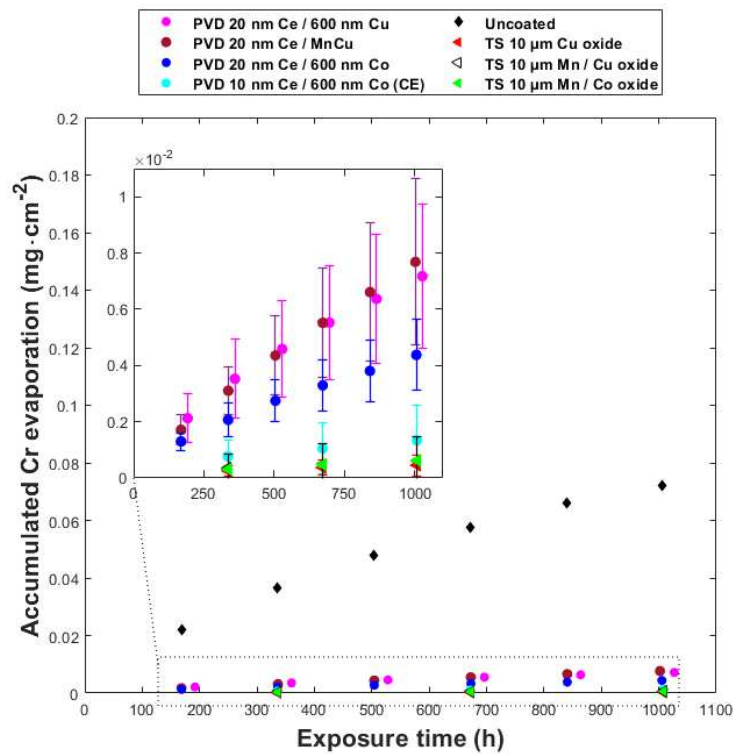


Figure 1: Cumulative Cr evaporation as a function of time for the PVD Ce/Cu-coated samples (magenta circles), PVD Ce/MnCu-coated samples (brown circles), PVD Ce/Co-coated samples (blue circles), PVD Ce/Co-coated samples with coated edges (cyan circles), TS Cu-coated samples (red triangles), TS Mn/Cu-coated samples (open triangles), TS Mn/Co-coated samples (green triangles), and uncoated samples (black diamonds) that were exposed for 1,000 h at 650°C in air that contained 3% water vapour. Error bars indicate the standard deviation.

The PVD Ce/Co and Ce/Cu coatings, as well as the TS Mn/Co coating exhibited the formation of thin protective Cr_2O_3 scales underneath the coating. These samples also exhibited the lowest area-specific resistance (ASR) values. The remainder of the samples exhibited much higher mass gains and higher ASR values. Cr(VI) evaporation measurements showed that all the coatings behaved approximately the same despite the PVD coatings being only about one-tenth of the thickness of the TS coatings. Table 1 summarizes the behavior of the different coatings.

Material	Deposition process	Coating		Efficiency
		Inner	Outer	
AISI 441	PVD	20 nm Ce	600 nm Cu	Very good
AISI 441	PVD	20 nm Ce	600 nm Mn/Cu	Bad
AISI 441	PVD	20 nm Ce	600 nm Co	Very good

AISI 441	TS	10 µm Cu oxide	Very bad
AISI 441	TS	10 µm Mn/Cu oxide	Bad
AISI 441	TS	10 µm Mn/Co oxide	Very good

Table 1: Comparison of the different coatings.

Overall, PVD Ce/Cu coating could be an excellent alternative to the state-of-the-art PVD Ce/Co coating, as it exhibits similar corrosion characteristics. The use of copper instead of cobalt would also reduce the production costs for interconnects.

Influence of Deformation

The interconnects used in solid oxide fuel cells (SOFC) are usually shaped into a corrugated form that creates gas channels, furthermore coatings are applied onto an interconnect to increase its longevity by reducing Cr(VI) evaporation and oxide scale growth. Nowadays the manufacturing of interconnects mostly remains a multiple-step batch process. First, the interconnect must be stamped into a manufacturer-specific design, and afterwards a coating is applied batchwise. Even though the coating composition varies from producer to producer, its function does not. Within the range of the most commonly used operating temperatures of SOFCs (600 °C - 850 °C), ferritic stainless steels used as an interconnect material corrode heavily. Coatings are applied onto the interconnect to avoid corrosion as well as related corrosion phenomena. The most commonly used coating is $(\text{Mn,Co})_3\text{O}_4$ (MCO). In contrast to the conventional coating process for which the oxide is directly applied onto the steel using powder-based methods, conversion coatings rely on coating the steel with a metal. This means that for the MCO coating, the Co is applied onto the interconnect, where it quickly oxidizes to form Co_3O_4 during high temperature exposure. Outward diffusion of Mn from the steel to the surface then leads to the formation of MCO. The great benefit of this coating route is that a large-scale roll-to-roll physical vapor deposition (PVD) process can be applied, in which large amounts of steel can be coated continuously. This manufacturing route has the added advantage that very thin, and highly dense coatings can be obtained, and no extra densification step is required, as is often the case for non-conversion coatings. However, if the interconnect is first stamped and then coated, the large-scale roll-to-roll process cannot be implemented. On the other hand, if the interconnect is first coated and then stamped (hereinafter called pre-coated), the coating will crack during deformation and this could lead to increased Cr evaporation or even delamination of the entire coating.

Earlier work has shown that Ce/Co-coated material that had been pre-coated and found that the cracks, which had formed in the coating during deformation, had completely healed after only 24 h of exposure at 850 °C. The cracked areas were initially covered by a $(\text{Cr,Mn})_3\text{O}_4$ layer. The self-healing was accomplished by the

fast lateral Co diffusion between these areas resulting in a homogenous $(\text{Co,Mn})_3\text{O}_4$ cap layer over the entire surface of the coated FSS. The Cr evaporation verified that the cracks healed extremely fast, and no significant difference in Cr evaporation was observed between the undeformed Ce/Co-coated FSS and the deformed Ce/Co-coated FSS. However, even though the process of self-healing occurs rapidly at 850 °C and therefore large-scale roll-to-roll manufacturing processes could be used at these operating temperatures, it remained very doubtful if the healing process also occurred at lower operating temperatures. This was mainly due to the fact that the self-healing mechanism relies heavily on lateral Co diffusion along the surface of the coated material. Data by Grzesik et al. suggests that the diffusion of Co ions is one to two orders of magnitude lower at 650 °C than at 850 °C. Therefore, the self-healing mechanism might be too slow at 650 °C to effectively inhibit Cr poisoning.

Figure 2a shows the results for AISI 441 exposed to air at 850 °C. As expected the Cr evaporation rates for Ce/Co-coated AISI 441 decreased by roughly a factor of 10 compared to the uncoated AISI 441, and the average rate was around $3.2 \cdot 10^{-5} \text{ mg cm}^{-2} \text{ h}^{-1}$. Undeformed and deformed Ce/Co-coated AISI 441 showed a significant scatter in Cr evaporation rate at 850 °C, but no clear trend could be detected between the deformed versus undeformed material. The Cr evaporation rate at 750 °C is shown in Figur 2b. The rate for uncoated AISI 441 at this temperature was on average $1.4 \cdot 10^{-4} \text{ mg cm}^{-2} \text{ h}^{-1}$. A slight difference in the Cr evaporation rate behavior was visible between the undeformed and deformed Ce/Co-coated AISI 441 that was exposed to air at 750 °C. The Cr evaporation rate was nearly constant with an average rate of $1.2 \cdot 10^{-5} \text{ mg cm}^{-2} \text{ h}^{-1}$ for the undeformed material. On the other hand, the initial average Cr evaporation rate for the deformed Ce/Co-coated AISI 441 was $2.1 \cdot 10^{-5} \text{ mg cm}^{-2} \text{ h}^{-1}$, however, this rate quickly decreased by a factor of 1.75, and both the deformed and undeformed Ce/Co-coated AISI 441 showed similar Cr evaporation rates after only 168 h.

Figure 2c and c1 depict the Cr evaporation rate for AISI 441 exposed to air at 650 °C. The Cr evaporation rate for the uncoated material decreased in the beginning, and a plateau seemed to be reached after more than 360 h of exposure. The rate after the decrease was around $7.8 \cdot 10^{-5} \text{ mg cm}^{-2} \text{ h}^{-1}$. Ce/Co-coated AISI 441 exhibited much lower Cr evaporation rates than the uncoated material. However, the initial rates for the deformed Ce/Co-coated AISI 441 were substantially higher ($2.6 \cdot 10^{-5} \text{ mg cm}^{-2} \text{ h}^{-1}$) than those for the undeformed Ce/Co-coated AISI 441 ($3.3 \cdot 10^{-6} \text{ mg cm}^{-2} \text{ h}^{-1}$). Nevertheless, after 360 h of exposure, the rates for the deformed material had decreased by a factor of 7.9 and had reached the same level as those for the undeformed material. At 650 °C Cr-evaporation measurements were also conducted on heat-treated deformed Ce/Co coated

AISI 441. The rates for these samples were similar to those of undeformed Ce/Co coated AISI 441 throughout the exposure, even in the initial phases.

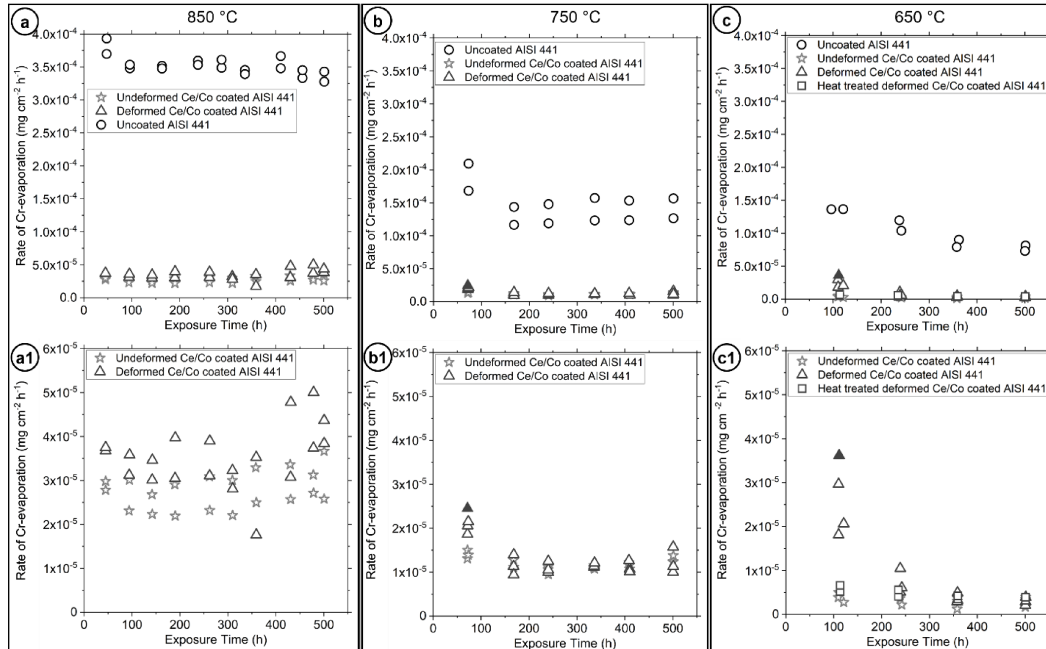


Figure 2 Cr evaporation rates for uncoated and Ce/Co-coated AISI 441. Samples were exposed to air at 850 °C (a and a1), 750 °C (b and b1), and 650 °C (c and c1). The bottom row depicts zoomed in versions of the top row. Filled symbols mark samples that were taken out after 71 h or 111 h of exposure at 750 °C or 650 °C, respectively.

The data in Figure 1 clearly indicate that the process of self-healing was significantly slower at 650 °C, and according to the Cr evaporation data the process seemed to be complete only after 360 h of exposure compared to few h at 850 °C. This is expected due to the slower kinetics but importantly even at 650 °C a complete healing occurs and after ~300 h of exposure no significant difference can be observed between deformed and undeformed material.

To further corroborate these findings the oxide scale of the deformed samples was investigated with Transmission Electron Microscopy (TEM) (see Figure 3.). The non-cracked region showed a pure Co oxide as the top layer, with a thickness of around 1.2 μm and only minor incorporation of Mn (around 2 cationic%) close to the innermost layer of this oxide. The Co signal for the cracked region suggests that small amounts of Co had diffused into the crack. A very thin Co oxide layer (marked as region I) had formed (~30 nm) on top of the crack, and up to 30 cationic% Co was found in this layer. More Mn, around 15 cationic%, seemed to be present in the

Co-rich oxide covering the cracked area compared to the one covering the non-cracked area. Additionally, high amounts of Fe, between 35 and 40 cationic%, were found in the Co-rich oxide, and increasing amounts of Cr were found in this oxide, from 5 cationic% in the outermost layer to 30 cationic% in the innermost layer. A Cr-rich oxide (marked as region II) had formed underneath the Co-rich oxide. The TEM investigation thus confirms the hypothesis established on the basis of the Cr evaporation data: Even at the low temperature of 650 °C Co diffusion is sufficiently fast to establish a continuous outermost cap layer that blocks Cr evaporation even though the sample has been deformed heavily.

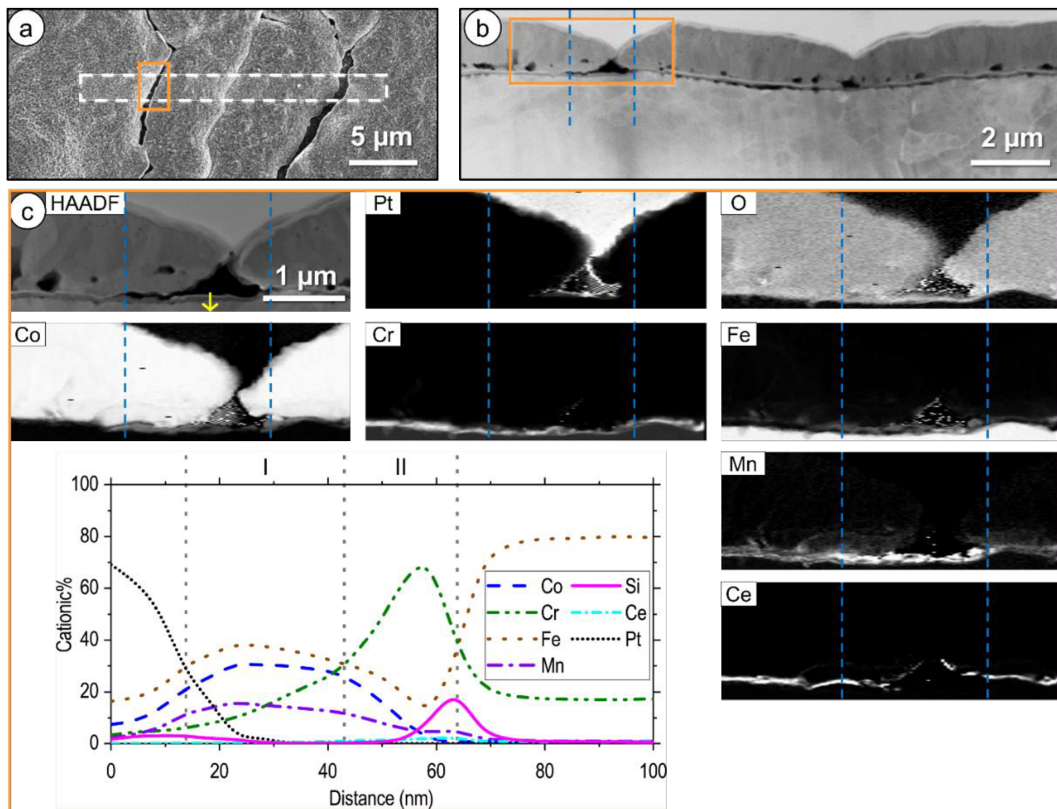


Figure 3: TEM images of the oxide scale of the biaxially deformed Ce/Co-coated AISI 441 that was exposed to air for 111 h at 650 °C. The area marked by the orange box was investigated with STEM/EDX and the blue lines mark the crack location. A) SEM micrograph with the area chosen for the FIB lift-out marked by the dashed white box; b) HAADF micrograph of lift-out; c) HAADF micrograph including the corresponding EDX maps in at% and a line scan through the crack (yellow arrow).

Dual atmosphere corrosion

Under fuel cell conditions, interconnects are exposed to two different atmospheres in parallel. One side of the interconnect is exposed to air (air-side), while the other side is exposed to H₂ (fuel-side). Many studies have focused on the effect of the dual atmosphere on the corrosion behavior of FFS interconnects. Most of them have found

increased corrosion on the air-side when hydrogen is used as the fuel. The effect of hydrogen is still unclear, and a lot of discrepancies have been noted. On the one hand, some studies of samples exposed to the dual atmosphere at high temperatures (around and above 800°C) have shown enrichment of Fe on the oxide scale or the formation of thicker oxide scales. The trend to lower the operational temperatures is expected to reduce material degradation, allowing for the use of cheaper materials. However, research at Chalmers has shown an increased dual-atmosphere effect at lower temperatures.

This inverse temperature effect, as described by Alnegren et al., i.e., a more pronounced effect observed at lower temperatures than at high temperatures has been attributed to a reduction of Cr diffusion from the bulk, causing a decrease in Cr at the metal/oxide interface and leading to the formation of break-away oxidation. Gunduz et al. recently concluded that hydrogen decreased the Cr diffusion at the grain boundaries, decreasing the Cr supply to the scale and thus promoting the formation of a non-protective Fe-based oxide scale. The severity of the effect can be observed in Figure 4a depicts the surface of uncoated 441 steel after 0h at 600h in dual atmosphere conditions. 0h means that the sample is ramped up to 600°C (1°/min) and immediately cooled down again (1°/min). It can be observed that almost 50% of the surface are covered with thick oxide islands consisting of an outer Fe₂O₃ oxide scale and an inner part that consists of Fe-Cr spinel. The oxide islands are about 20 µm thick and this catastrophic behavior is termed breakaway corrosion.

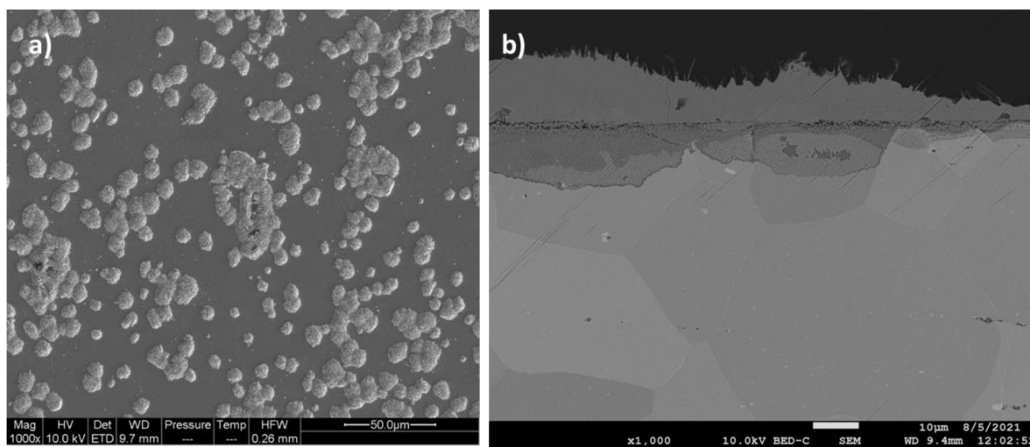


Figure 4: a) SEM top view image of uncoated 441 after 0h of dual atmosphere exposure at 600°C. b) typical breakaway morphology with an outward growing Fe₂O₃ oxide and an inward growing Fe-Cr spinel.

In order to mitigate the dual atmosphere effect different mitigation strategies have been tested. Three approaches have proven particularly successful. A) cold working the uncoated steel. Figure 5a shows an SEM cross-section of uncoated 441 and ground (P1200 grit) steel exposed under identical dual atmosphere conditions as above but for 336 h. As can be seen in the image the grinding causes the formation

of a 2-3 μm deep recrystallization layer that promotes Cr transport to the surface. The oxide scale on the sample is barely visible ($\sim 100\text{nm}$) and is a clear testament to this vast improvement in lifetime. Figure 5b and c depict a sample that has been coated with the above-described Co/Ce coating and subsequently pre-oxidized for 5h at 800 $^{\circ}\text{C}$. The images have been taken after 3000h of dual atmosphere exposure. Although the SEM image (Figure 5c) shows the formation of some large oxide nodules these barely grow in size and it depicts a dramatic improvement compared to the 0h experiment depicted in Figure 5.

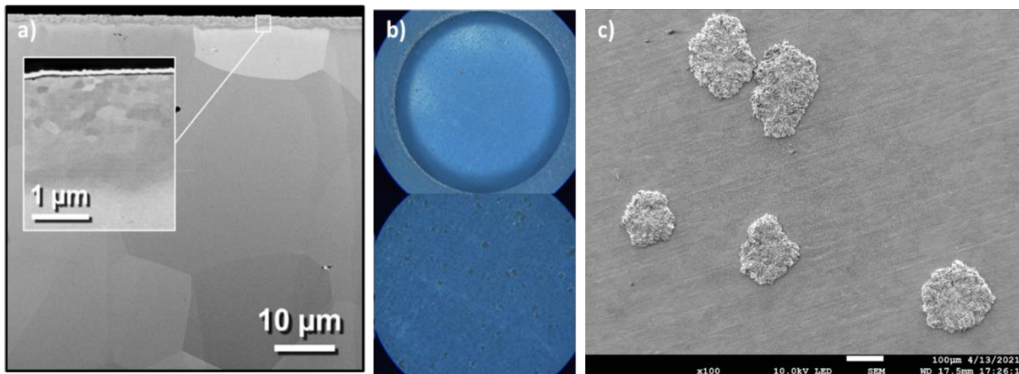


Figure 5: uncoated 441 steel after exposure under dual atmosphere conditions. a) uncoated material that was ground prior to oxidation after 336h at 600 $^{\circ}\text{C}$, b) optical image of Co/Ce coated 441 after 3000h of exposure c) SEM image of the surface of the same sample as in b)

Stack tests

Elcogen was responsible for stack assembly and testing phases. Two stacks were assembled during to project, where the main idea was to study the properties of two different interconnect materials a) Crofer 22 APU and b) AISI 441, coated with two different coating technologies: 1) Elcogen's $(\text{Mn,Co})\text{O}_4$ coating and 2) Sandvik Materials Technology (SMT) Ce/Co coating to prevent evaporation of the Cr-species which have shown to cause increased stack degradation.

Elcogen has carried out multiple stack tests where Crofer 22 APU is used together MCO coating. The longest test with this material system has lasted over 25,000 hours (Figure 6) and therefore this was selected to be a reference material combination in this project. This long-term test showed extremely low degradation rate (0.4%/1000 h) which indicated that 40,000 hour life-time can be achievable.

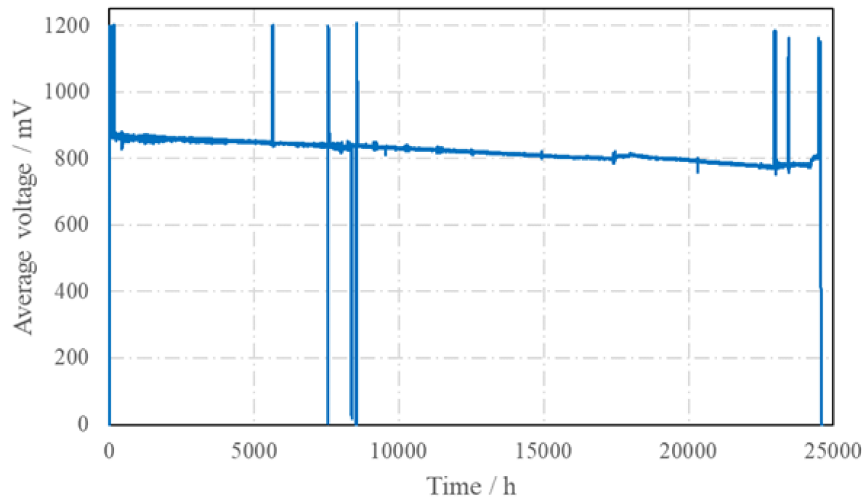


Figure 6. Standard ElcoStack E350 operated at 25,000 hours.

The stacks were tested in collaboration with VTT Technical Research Centre of Finland (VTT), which have a possibility operate a dual-stack test station (Figure 3), meaning that both stacks were tested at the same time. The advantage of this station is that both stacks are experiencing the same temperature and having the equal gas composition and flow rates, minimising the errors when interpreting the data. The stack test lasted in total of 3300 hours. After the test was over, both stacks were disassembled and selected ICs were used for the post-mortem analyses.



Current @ NOC	30 A
Fuel composition	50:50 H ₂ /N ₂
Fuel Flow	15.5 I _N /min
Air Flow	33 I _N /min
FU @ NOC	40%
Fuel Inlet Temperature	620°C
Air Inlet Temperature	620°C

Figure 7. Dual-stack test station in VTT with installed E350 stacks and operating parameters

The stack, assembled by using Crofer 22 APU as an interconnector, showed typical degradation profile and rate (Figure 7). The voltage curves (left) represent the average cell voltage values for each coating technology and columns (right), degradation rates for individual cells after a certain number of hours. Typically, the

cell voltages are increasing at the beginning of the operation as the electrical contacts are improving and the electrochemically active sites are activating. Once this activation period is over, the stack degradation is determined by growth of the oxide scales, material diffusion to the electrolyte layer, fuel electrode coarsening and Cr-poisoning of the air electrode. All of these ageing processes are causing steady voltage drop. As seen from the columns, the fastest degradation rate increase is measured during the first 2000 hours. This indicates that the predominant degradation mechanisms may include fuel electrode deactivation, oxide scale growth on the metal surfaces or interdiffusion processes between the air electrode and the electrolyte.

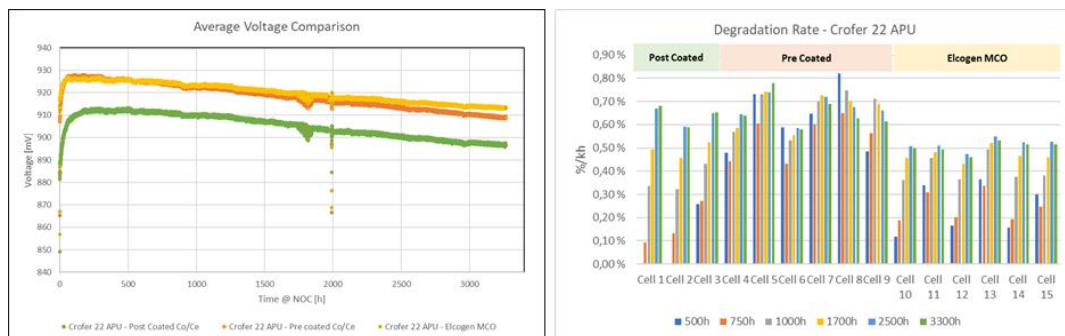


Figure 8. Average cell voltages (left) and degradation rates with specific intervals (right) measured for cells assembled by using coated Crofer 22 APU interconnectors

The degradation rate from 0.40 to 0.52 %/1000h, obtained for Elcogen MCO coated Crofer 22 APU is close to values reported to 25,000 hour stack, proving that the stack test has been valid. Pre- and post-coated Ce/Co showed slightly higher (0.70 %/1000h) degradation rate after 3300 hours. The thickness of the MCO coating was 10 μm , whereas Ce/Co coatings were less than 1 μm , meaning that MCO coating may provide lower Cr evaporation rate and/or reduced chromia growth rate on the steel surface compared to Ce/Co coating. The difference in degradation rate between pre- and post-coated Ce/Co protective coatings is marginal indicating stable oxide layer formation in both coating variants even though pre-coated IC have cracks on the coating due to forming process.

Figure 9 shows the average cell voltage profile and degradation values for the unit layers assembled by using AISI 441 interconnectors. There is an obvious difference in the voltage behaviour compared to the stack made from Crofer 22 APU and is highlighted after 200 hours.

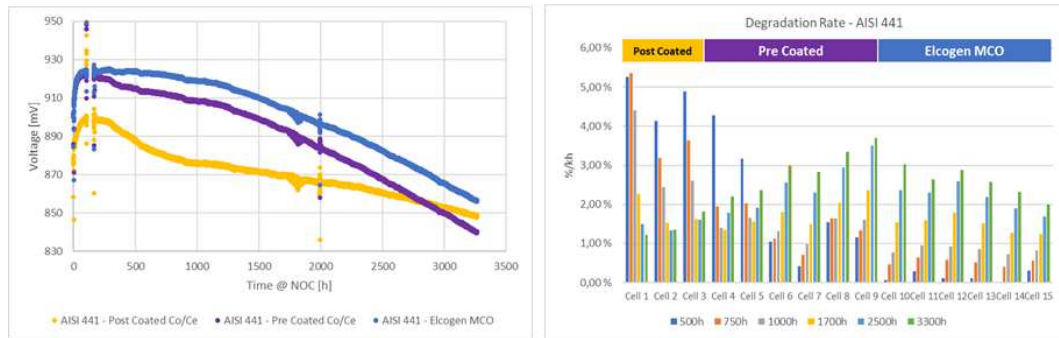


Figure 9. Average cell voltages (left) and degradation rates with specific intervals (right) measured for cells assembled by using coated AISI 441 interconnectors.

According to the results, AISI 441 gives higher degradation rate than Crofer 22 APU. Only the-post coated Ce/Co AISI 441 showed some signs of saturation in degradation behaviour. However, Elcogen MCO and Sandvik pre-coated Ce/Co showed increasing degradation rate throughout the test campaign, exceeding values of 2.0 %/1000h and 4.5 %/1000h, respectively. Typically, the higher degradation is connected to the cathode side oxide growth and Cr-poisoning. However, when doing a post-mortem disassembly, it was noticed that; 1) the anode side had an oxide scale with dark green colour and 2) the cell had poor adhesion with the ICs. The green colour can be linked the presence of pure Cr-oxide. Cr-oxide has a poor electrical conductivity compared, for example, for MnCr-oxide with is typically observed when Crofer 22 APU is used as interconnector.

The 3000 hour stack test showed that Sandvik's Ce/Co PVD coating can be suitable coating alternative to decrease degradation mechanisms. The best results were obtained by using Crofer 22 APU, with slightly higher degradation rate compared to Elcogen standard MCO coating. It is clear that degradation rates increased when AISI 441 was used as interconnectors. The same increase in degradation rates were observed regarding which coating technologies were used. This indicates that degradation mechanisms are also linked to the anode side, which was not coated in these studies. There are several ways to address anode side degradation mechanisms: 1) optimise the stack conditioning process so that the growth of the anode side oxide scales is decreased, 2) optimise the conditioning process so that oxide scale composition is more conductive and/or 3) apply a coating on the anode side which decreases the growth of the oxide scale and improves the adhesion between the ceramic cell and metallic interconnector.

Diskussion

The project resulted in significant progress in developing a low-cost interconnect solution for SOFC. Sandvik's world unique pre coated Co/Ce coated material was successfully integrated into a stack from Elcogen and operated for 3000h. The degradation rate is in the same range as the Elcogens state of the art (SoA) solution. This is major breakthrough and a proof of concept for the use of pre-coated steel in SOFC stacks. The pre coated steel concept allows for high volume production in a

very cost-effective way. The results obtained in this project will facilitate the commercialization of SOFC technology to a broader market by increasing the lifetime and decreasing the cost of SOFC stacks.

In the project we also developed an alternative approach using custom-made coatings for low-cost steels. Focus was on the low cost steel AISI 441. The coatings evaluated included Cu/Ce and Cu/Mn/Ce as well as including Co/Ce and Elcogens state of the art coating. The coatings were tested both in lab-scale and in stack tests. The laboratory results were very promising, Co/Ce coated 441 performed extremely well in experiments for up to 40 000h. The material showed very slow corrosion rates, no sign of degradation of the coating and low Area Specific Resistance (ASR). A reduction of chromium evaporation by up to 98% using Co/Ce and Co/Cu coatings were obtained. The life time of the coated steel could also be extended up to 20 times longer compared to uncoated steel. The implementation of low cost AISI 441 into a stack showed however that the degradation rate was much higher than the more expensive special steel (Crofer 22 APU.) Further research is needed in order to overcome this, one approach would be to, in addition to cathode side coatings, also implement anode side coatings.

Publikationslista

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Bilagor

- Administrativ bilaga (se mall)
- Vetenskapliga artiklar (färdiga, accepterade, eller inskickade manus)
- Avhandlingar