SOFC STACK DEVELOPMENT
AT UNITED TECHNOLOGIES CORPORATION

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ABSTRACT

United Technologies Corporation is developing a low-cost solid oxide fuel cell stack technology that offers long-life durability and thermal cycling robustness for building cooling heating and power applications. Our design philosophy decouples the coefficient of thermal expansion and oxidation requirements for metallic interconnects, enabling us to use oxidation resistant nickel-based superalloy interconnects. Innovative interconnect designs along with non-contaminating seals offer tremendous advantages in producing long-life, thermal cycle tolerant stacks. 1-cell and 3-cell stack tests for up to 4500 hours and 20 thermal cycles show excellent stack durability.

INTRODUCTION

United Technologies Corporation (UTC) has been developing fuel cells for the last 40 years. UTC’s alkali fuel cells power the space shuttle and have so far accumulated 113 missions and over 90,000 hours. UTC’s PureCell 200 (formerly called PC-25) phosphoric acid fuel cell systems have been in operation since 1991 and have accumulated over 6 million operating hours. Solid oxide fuel cell (SOFC) technology offers opportunities for high efficiency and low cost electricity generation coupled with high quality waste heat. These features make SOFC prime movers highly desirable for building cooling, heating and power (BCHP) applications. Over the past few years, UTC has been involved in developing SOFC stacks with the objective of meeting durability and cost targets required for BCHP applications. Our principal development considerations have been long life >40,000 hours (preferably >60,000 hours) and low cost, so as to be competitive with existing reciprocating engines and developing microturbines. Thermal cycling requirements are moderate with several hundred thermal cycles anticipated over the life of the stack for medium and large buildings.

DESIGN PHILOSOPHY

Our approach at United Technologies Research Center (UTRC) has been based on planar anode-supported cells with metallic interconnects, using oxidation resistant alloys that meet the life requirements. Planar anode supported cells offer high power densities, modest footprint size and adequate robustness that are envisioned to translate into low cost, reliable stacks that will meet BCHP requirements. Interconnect oxidation and
sealing have been the two greatest problems with planar SOFCs using metallic interconnects. The SOFC community has mainly focused on Fe-Cr alloys (also known as ferritic stainless steels) as the interconnect material. While ferritic stainless steels possess a good match in coefficient of thermal expansion (CTE) with the cell, oxidation data suggests that these materials are unlikely to meet required lifetimes. On the other hand, oxidation resistant nickel-based super alloys such as Haynes 230 have shown the capability to meet life requirements. While these superalloys have demonstrated exceptional oxidation resistance, much of the SOFC community has not tried to use these materials in SOFC stacks, principally due to their higher CTE compared to the cells.

Sealing has been a major issue with planar anode supported cells. Glasses, glass-ceramics and mica have been explored extensively. There is substantial concern regarding life of these types of seals under thermal cycling as well as long-term exposure to gas streams laden with water vapor.

We have developed a unique set of cell-stack materials and design solutions using interconnects and seals that offer improved steady state durability while minimizing thermal stresses, leading to cell-stacks with good operational durability and robustness to thermal cycling. In addition, our design solutions enable increased in-plane footprint scale-up, leading to lower cost cell-stacks.

OXIDE SCALE GROWTH AND AREA SPECIFIC RESISTANCE

Figure 1 is a compilation of oxidation test results for ferritic stainless steel (E-BRITE) and nickel-based superalloy (Haynes 230) (1, 2). While there is substantial scatter in the data, possibly due to variations in surface and test conditions, Haynes 230 demonstrates better oxidation resistance than E-BRITE. The oxidation rate constant for E-BRITE is >10x higher than Haynes 230 at temperatures between 700 and 800°C. Figure 2 shows our oxidation data for 10,000 hours followed by parabolic scale growth projection to 40,000 hours for tests conducted at 750°C in still lab air. Our 40,000 hours projection for oxide scale growth is about 1.5 μm for Haynes 230 and 4.1 μm for E-BRITE. Electrical measurements conducted at UTRC using accelerated testing show that area specific resistance (ASR) values <0.1 Ω-cm² can be achieved after 40,000 hours at 750°C using Haynes 230 interconnects. Thicker but more conductive scales that are formed on some novel ferritic stainless steels are prone to thermal cycling spallation and are unlikely to meet BCHP stack durability requirements. In addition some thin foil based bipolar plate designs using ferritic steels are prone to perforation by localized oxidation during long-term stack operation, resulting in cross over and cell degradation.

INTERCONNECT DESIGN

The interconnect-cathode interface is one of the weakest locations in an SOFC stack and is prone to damage during thermal cycling. Our interconnect design approach minimizes CTE mismatch induced thermal stresses that are generated between the interconnect and the cathode. Numerical simulations show >3x reduction in stresses at the interconnect-cathode interface with UTC’s interconnects compared to conventional CTE matched ferritic stainless steel interconnects. In addition, nickel-based superalloys offer
unparalleled advantage compared to ferritic steels in creep resistance. For example, 1% creep strain in 40,000 hours requires a stress of 30 MPa for Haynes 230 (3) vs. 2 MPa for E-BRITE (4). Our design options decouple the CTE and oxidation requirements for metallic interconnects and offer tremendous advantages in producing long-life, durable stacks.

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log \( k_p = -10.839x + 0.6293 \)

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-10.839x + 0.6293

-12

log \( k_p = -7.9625x - 2.4762 \)

log \( k_p = -18.349x + 5.9686 \)

1000/T (1/K)

Figure 1. Arrhenius rate constants for oxide scale growth.

Figure 2. Chromia scale growth with Haynes 230 and E-BRITE.

SEAL DESIGN

Our compressive seals have been designed using materials that do not contaminate the cell. These seals are configured to produce non-hermetic but adequate sealing under a small compressive load. Seal testing demonstrates minimal change in leak rate under...
steady state operation and thermal cycling. Figure 3 shows seal testing data before and after thermal cycling demonstrating about 1% leak rate using He.

![Graph showing seal leak rate before and after thermal cycling.](image)

**Figure 3.** Seal leak rate before and after thermal cycling.

**CHROMIA POISONING MITIGATION**

Chromia poisoning of the cathode is a major degradation issue with common electrodes. Cathodes such as strontium doped lanthanum manganite (LSM) and strontium and cobalt doped lanthanum ferrite (LSCF) will degrade significantly in a chromia environment. We have developed approaches to mitigate chromia poisoning through chromia containment by means of application of cost effective coatings. Figure 4 shows an ongoing single cell stack test with <1%/1000 hours iR-free degradation over 600 hours of testing using this approach.

![Graph showing steady state test with chromia poisoning mitigation strategy.](image)

**Figure 4.** Steady state test with chromia poisoning mitigation strategy.

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ELECTROCHEMICAL TESTING

We have been testing cells of different size and different levels of sub-component level integration and electrochemical diagnostics – ranging from sub-scale single cells in a "clean" environment to full scale multi-cell stacks in simulated environments. Electrochemical diagnostics include current interruption, electrochemical impedance spectroscopy and multiple voltage sensing. "Clean" environment tests have been used to determine intrinsic stability of the cells as well as to understand the effect of contaminants under accelerated testing conditions. Figure 5 shows a plot of an LSM cathode cell tested in a "clean" environment showing <0.2%/1000 hours degradation in tests conducted for 400 hours.

![Figure 5. Single-cell test conducted in a “clean” environment.](image)

STACK TESTING

Three-cell stack tests were conducted for up to 4500 hr (Figure 6). Results show 1.2 to 1.5%/1000 hr steady-state degradation. Further improvement is required for BCHP applications. Results are based on non-optimized chromia poisoning mitigation strategy and older generation of cells. As discussed, newer tests show <1%/1000 hr steady-state degradation in a chromia environment. Figure 7 shows thermal cycling data from a 1-cell stack. Minimal degradation was observed after 15 thermal cycles (750° to 100°C) in these tests. Ongoing 1- and 3-cell stack tests with improved interconnect design show excellent response to thermal cycling. Future activities are scaling up to a 1-kW stack, root cause analysis, and continuous improvement of performance and durability.

CONCLUSIONS

United Technologies Corporation is developing a low-cost, durable solid oxide fuel cell stack technology aimed at building cooling heating and power applications. Our design philosophy decouples coefficient of thermal expansion and oxidation resistance requirements, enabling us to use oxidation resistant nickel based superalloys. Short stack tests demonstrate excellent steady state and thermal cycling durability.
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