

Roll-to-Roll SOFC Manufacturing for Low Cost Energy Generation



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Roll to Roll SOFC Manufacturing for Low Cost Energy Generation

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ACRONYMS

AFL	Anode Functional Layer
AMO	Advanced Manufacturing Office
ASL	Anode Support Layer
ASR	Area-Specific Resistance
BMF	Battery Manufacturing Facility
CRADA	Cooperative Research and Development Agreement
DFMA	Design for Manufacturing and Assembly
DOE	Department of Energy
EERE	Energy Efficiency and Renewable Energy
GDC	Gadolinium Doped Ceria
OCV	Open Circuit Voltage
ORNL	Oak Ridge National Laboratory
R2R	Roll-to-Roll
SBV	Small Business Vouchers
SOFC	Solid Oxide Fuel Cell
TEA	Techno-Economic Analysis

EXECUTIVE SUMMARY

Oak Ridge National Laboratory (ORNL) and Redox Power Systems, LLC. collaborated on this project to develop an integrated, continuous roll-to-roll (R2R) manufacturing processes to produce solid oxide fuel cells (SOFCs). SOFCs are ceramic-based electrochemical devices that efficiently produce electricity from hydrogen-containing fuels, including natural gas. Standard processes for manufacturing SOFCs include tape casting of the anode and electrolyte, followed by stacking of the layers, and lamination using a uniaxial press or isostatic press. After high temperature firing, the cathode and electrical contact are screen printed and fired once again to complete the cell.

This effort aimed to further develop R2R tape fabrication processes for green cell fabrication with the DOE Battery Manufacturing Facility (BMF) at ORNL as next generation improvements to current methods, focusing on R2R calendaring and slot die coating for lower cost, high volume approaches for multilayer green cell fabrication. Two paths were investigated in parallel in this project. The first path was to optimize R2R processing using traditional tape cast materials from the Redox production chain on the BMF's heated calender for compressing and laminating multilayer materials. The second path was to use BMF's pilot-scale slot-die coater for R2R manufacturing as an alternative to tape casting of some or all of the layers in the green cell. Processes have been developed and and process conditions have been identified. Additionally, basic process economics were estimated for comparison to Redox current manufacturing approaches for the applicable process modules. The SBV program results will enable Redox to advance lower cost R2R manufacturing processes to produce millions of green cells per year on a single production line.

As a result of this work, one invention disclosure (Invention Disclosure ID#: 201804281, "Roll to roll SOFC Manufacturing for low cost energy generation" has been submitted. In addition, 1 paper is under preparation for submission to a peer-reviewed journal.

1. OBJECTIVE

Redox develops high-performance solid-oxide fuel cells (SOFCs) for distributed-generation power systems, using high power density, intermediate-temperature cell technology. The high power density and intermediate operating temperature of Redox's SOFCs results in lower costs and more compact generation systems compared to incumbent SOFC technologies. Redox SOFCs can be tailored to operate under various conditions, including with a variety of hydrocarbon fuels, at a range of operating temperatures, with a variety of load profiles, and in combined heat and power applications.

Cell and stack components are responsible for between 40 to 60 percent of final system costs and are key to performance. R2R processing is a potential path to significantly increase Redox's production cell capacity to hundreds of megawatts per year, allowing the company to support mass markets faster and at lower costs than traditional manufacturing approaches.

ORNL is the largest multi-disciplinary Department of Energy (DOE) research laboratory with extensive expertise and capability in materials science and processing science. ORNL utilized Redox's recipes to develop R2R manufacturing processes for fabricating individual cell component green tapes through slot die coating and laminating multiple cell components into cell stacks, which will increase scale, improve yield, and dramatically reduce SOFC costs for mass markets.

2. INTRODUCTION

This project targeted the development of R2R manufacturing processes to produce SOFCs. This effort focused on further developing R2R tape fabrication processes for green cell fabrication with the BMF at ORNL as next generation improvements to current methods, focused on R2R calendering and slot-die coating for lower cost, high volume approaches for multilayer green cell fabrication. This project proceeded in two parallel paths. The first path was to optimize R2R processing using traditional tape cast materials from the Redox production chain on the BMF's heated calender for compressing and laminating multilayer materials. The second path was to use BMF's two pilot-scale slot-die coaters for R2R manufacturing as an alternative to tape casting of some or all of the layers in the green cell. ORNL and Redox collaborated closely during the project. ORNL led process development and optimization at the BMF. Redox led electrochemical characterization, microstructural characterization, and techno-economic modeling. The SBV program enables Redox to advance lower cost R2R manufacturing processes for future production of millions of green cells per year on a single production line.

3. BENEFITS TO THE FUNDING DOE OFFICE'S MISSION

The work performed under this CRADA directly supported DOE's mission in the SBV, part of the Technology-to-Market program within the U.S. DOE's Office of Energy Efficiency and Renewable Energy (EERE), which provides small clean energy businesses access to DOE's national labs' capability—making the contracting process simple, lab practices transparent, and access to the labs' unique facilities affordable. This project was in collaboration between Oak Ridge National Laboratory and Redox Systems, Inc. It focused on developing R2R processes for manufacturing of SOFCs, which significantly reduced manufacturing cost. The project results enable Redox to accelerate production rates by using R2R processing; enable US-based cell production to easily support mass markets; and leverage considerable US-based experience in high-yield R2R processing.

4. TECHNICAL DISCUSSION OF WORK PERFORMED BY ALL PARTIES

4.1 R2R CALENDERING PROCESS DEVELOPMENT

As mentioned above, the SOFC typically consists of multiple layers of different compositions combined together to form a cell, or laminate. For example, the GEN-1 Redox cell consists of (in the order listed) an anode support layer (ASL), an anode functional layer (AFL), an electrolyte layer, and a cathode layer. A conventional approach to manufacturing involves laminating layers of the ASL, AFL, and electrolyte together, followed by co-sintering at high temperature ($> 1300\text{ }^{\circ}\text{C}$, to ensure densification). After sintering, an electrode is typically screen printed on the sintered half-cell and then sintered at a relatively low temperature ($< 1200\text{ }^{\circ}\text{C}$, to retain porosity). The process of laminating layers is typically performed by cutting tape cast layers of each component (sheeting) and pressing them together one cell at a time (lamination), leading to a bottle-neck in production rate. In this project, we examined an approach to alleviate the bottle-neck using a roll-to-roll (R2R) process that brings together different sheets of material stored on rolls, compresses them together, and creates a new roll of the laminate in a continuous process. Since the R2R process can be very high throughput, thus consuming very large quantities of material, in this evaluation work layers were pre-cut and hand-fed through hot rollers to replicate the R2R process. Future work will involve scaling up the process developed in this program.

In the first stage of the calendering process development, commercial tape cast layers of ASL, AFL, and electrolyte layers were laminated together, where the layers consist of flexible binders holding together oxides of NiO-cermet (in the ASL and AFL) or gadolinium doped cerium oxide (GDC, in the electrolyte). Heat and pressure during calendering cause the binders to become sticky, binding the layers together. Redox provided the commercial tapes and, after calendaring at ORNL, sintered the laminates and performed metrology and tested them as operating SOFCs, as described in the next section. In the second stage, commercial tapes and slot-die coated materials were calendered, as described in a later section.

Lamination was carried out and optimized through the control of three lamination parameters, temperature, speed and gap. Lamination conditions were identified for two electrode layer components (i.e., ASL-ASL, ASL-AFL, and AFL-GDC), three electrode layer components (i.e.,

ASL-ASL-AFL), and four electrode layer components (i.e., ASL-ASL-AFL-GDC) to create an operation diagram for each configuration. A constant line speed was chosen for lamination of the four components since the lamination speeds should be the same in the actual end-to-end continuous process, while temperature and gap can be varied for each step. Dependence of lamination conditions on sample size was also investigated. Eventually, samples in 6 in x 6 in dimensions were laminated. 6 samples were laminated with three gaps of the calender press for the ASL-ASL-AFL part of the lamination process, referred to as large, medium and small herein and in Section 4.3.

Figure 1 shows the images of the 3 and 4-layer laminates. For the 3-layer laminates, the images were taken from the 2nd AFL layer side, which was laminated to a pre-laminated ASL-AFL. All three samples seem well laminated except some corner areas where there was mismatch in different layers due to differences in size and orientation during calendering. Each image shows the tan color of the electrolyte layer. Lighter colored regions, e.g., near the sample edges and circular shapes within the sample area indicate areas where the GDC electrolyte did not adhere well to the underlying AFL. Each sample exhibits some regions of poor electrolyte adhesion, but the majority of the sample is well adhered on visual inspection. There is no clear trend in laminate quality with calender gap setting. The amount of thickness compression during the calender process for the ASL-ASL-AFL did show an increase in compression with decrease in gap size, as expected (left axis in Figure 2). The final thickness after calendering all the four layers together, using the same gap in the additional calendering steps for each sample, did not show a significant trend with gap size for the ASL-ASL-AFL step, as shown in the right axis of Figure 2. The limited impact on gap size on final thickness likely arises from larger variations during lamination for the manual multi-lamination process and variations in casted tape thickness. In a fully operational, automated process, even tighter control over these variations would be expected. Overall, the calendering process resulted in successfully laminated samples that, as described in the next section, yielded testable SOFCs.

Processing the thick layers of ASL was difficult, often resulting in premature and thus undesirable release of the ASL from the carrier film during calendering. This behavior would serve as a large impediment to R2R processing, especially when rolling up the final thick laminated layers. In future work, carrier films with increased levels of “stickiness” would be investigated

(commonly controlled by the type of silicone based release agent applied to the carrier film) and modifications to the tape cast recipe to increase tape flexibility.

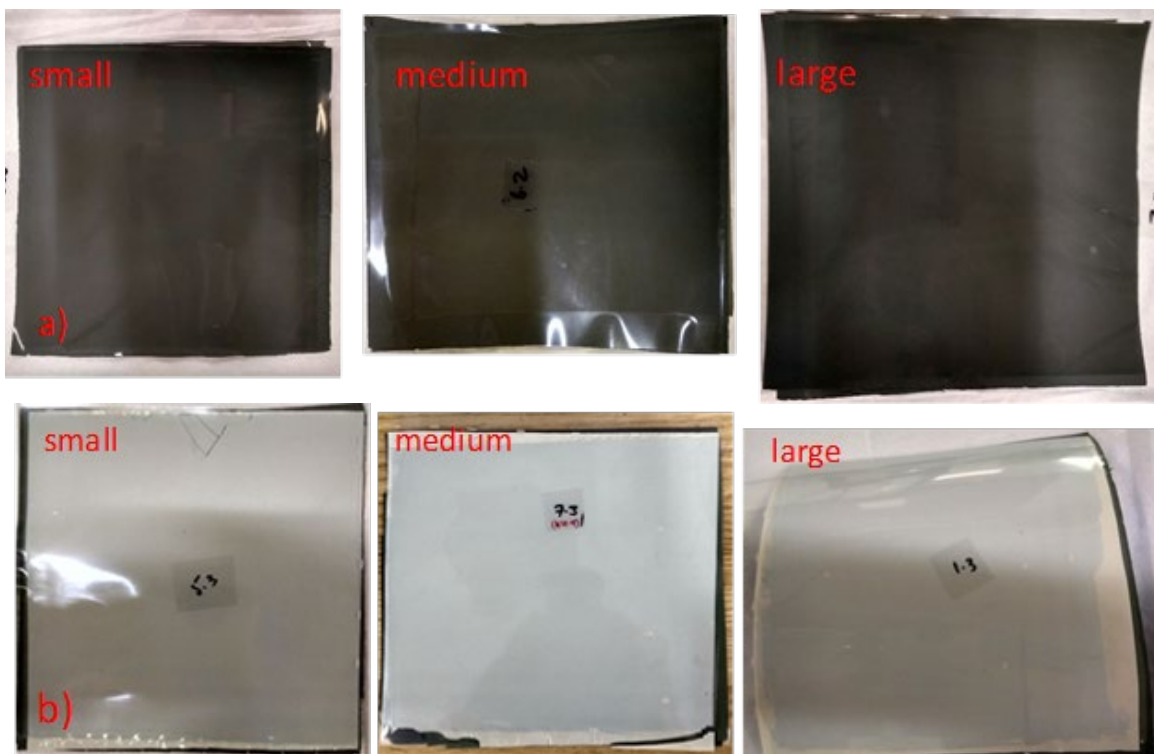


Figure 1. Images of the 3-layer (a, ASL-ASL-AFL) and 4-layer (ASL-ASL-AFL-GDC) samples (6 in x 6 in) laminated under small, medium and large gaps, respectively.

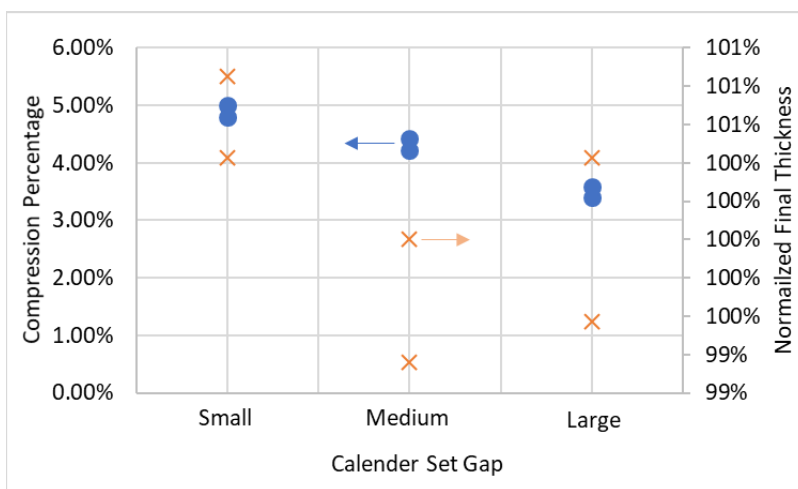


Figure 2. Compression percentage for the AFL-ASL-ASL calender step (blue circles) and normalized final thickness of laminates after all calender steps plotted against hot roller gap for the AFL-ASL-ASL calender step.

4.2 R2R SLOT-DIE COATING PROCESS DEVELOPMENT

R2R processes were developed to fabricate GDC, GDC on AFL, and AFL on GDC via slot-die coating. Slot-die slurry recipes were provided by Redox with minor modification made at ORNL. The slurry was mixed in a 1,000-ml bottle by adding the dispersant, GDC (or NiO-cermet), organic solvent, and zirconia milling media and ball milling overnight. Then binder and plasticizer was added and the solution was ball milled overnight again and then coated the following morning. In between ball milling and slot die coating there was a thirty-minute de-gas step in a planetary mixer. The slurry was then filtered through a discharge filtration system directly into the slot-die pump cart. A stainless-steel filter was used for the filtration. GDC, GDC on AFL, and AFL on GDC layers were successfully coated as shown in Figure 3, Figure 4, and Figure 5.



Figure 3. Images of slot-die coated GDC on mylar.



Figure 4 Images of GDC coating on slot-die coated AFL on mylar.

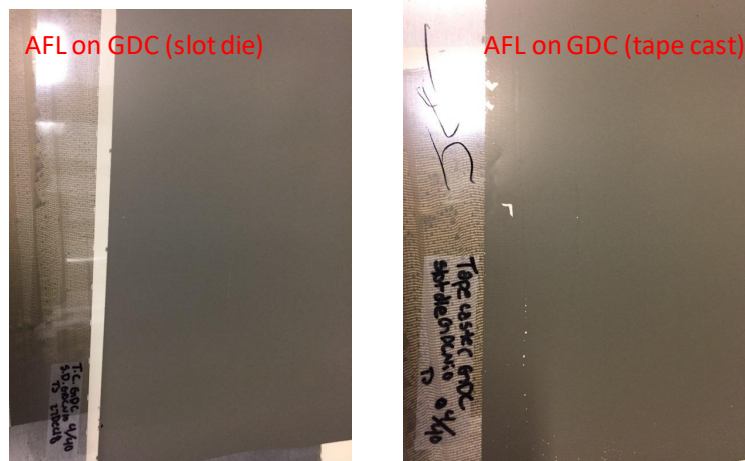


Figure 5. Image of AFL on GDC both fabricated from slot-die or tape cast where the AFL and GDC thicknesses are roughly the same.

4.3 SOFC CELL FABRICATION AND ELECTROCHEMICAL EVALUATION OF R2R MATERIALS

Calendered laminates of commercial tapes were sintered at Redox, evaluated with metrology techniques, and tested as SOFCs. Three different cell configurations were measured, labeled large, medium, and small gap which designates the relative gap width between the hot rollers during AFL-ASL-ASL calendering. Figure 6 shows the optical profilometry of two sintered half-cells, one processed with the large gap setting, the other with a small gap setting in the calendering process. The size of the samples is approximately 12 cm by 12 cm. Both samples exhibited vertical and horizontal lines which likely originate from discontinuities in the manual feeding operation. There is no ability to hold the carrier layer in constant tension for the small samples here, resulting in inconsistent feeding and loading rates of laminates into the rollers. There appears to be fewer defects for the small gap case, indicating this is a preferred option to avoid such defects. The lower defect count in the small gap case may be due to a more consistent feed from the greater force exerted on the laminates by the rollers. Several large pits in the upper right of the small gap case were observed, though their origin is less clear. For SOFC testing, cells that were 5 cm by 5 cm or 4 cm by 4 cm were cut from these larger samples.

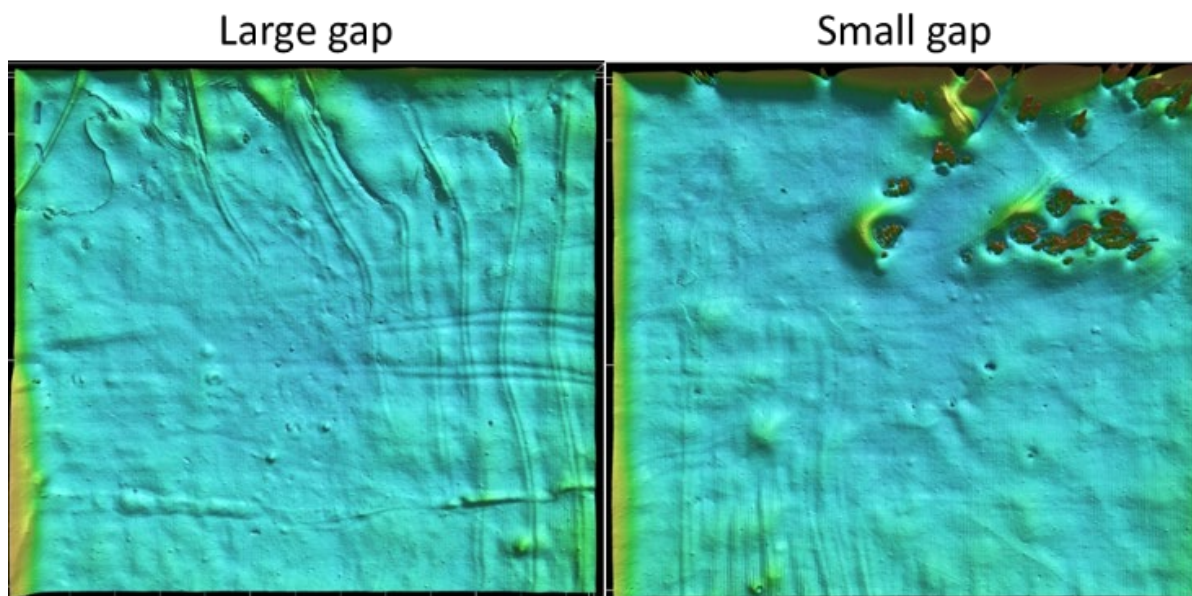


Figure 6. Optical profilometry (magnified 9 kx in out-of-plane direction to enhance defect detection) results of the electrolyte side of two sintered half-cells fabricated by calendering of commercial tape. Vertical and horizontal line defects, likely arising from hand-feed operation, are less prominent in the small gap as compared to large gap process conditions. The medium gap cell is not shown due to a sample loading error during the imaging process.

In the medium gap laminate, clear evidence of delamination of the ASL layers during sintering was observed after cutting the sintered sample. Figure 7 shows a photograph of the edge of one of the middle gap samples after cutting showing two layers that are separated. The relatively large thickness of the layers indicates that the two layers that separated were two ASL layers used in fabricating the laminate as other layers are much thinner. The layer separation was not observed prior to cutting the sample, indicating that lamination may not be fully uniform within tested samples. A more detailed post-test analysis is needed to fully characterize the extent of delamination defects. Besides working with even thinner roller gaps (which for too thin a distance may deform the laminate an unacceptable amount), improvements to the binder composition of the ASL tape (e.g., “stickiness”) are expected to increase lamination uniformity. For the SOFC testing described below, a cell that was cut from the same sintered samples which did not exhibit any obvious delamination was tested.

Optical images and profilometry of the SOFC tested cells are shown for the medium (Figure 8), small (Figure 9) and large (Figure 10) gap samples. In each case some discolorations can be observed on the surface, often with corresponding defects in the height profile. Note that

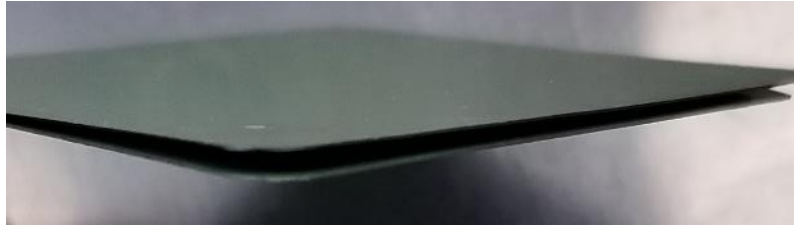


Figure 7. Photograph of the edge of one of the samples cut from the medium gap sintered cell showing two layers delaminated during sintering.

the range of heights shown for each height profile are the same, and the medium gap sample is a 4 cm by 4 cm sample, while the small and large gap samples are 5 cm by 5 cm samples. Each sample exhibits some impressions on the surface, with the vertical line processing defects most visible for the small gap sample. The small and large gap samples appear to generally suffer from the largest number of defects, though, as will be shown later, the small and medium gap samples exhibit the best SOFC electrochemical performance, indicating robustness against observable defects. It is also worth noting that while these cells exhibit a greater level of defects, as observed by the metrology here, the defect concentration and type (except for the line defects) is not exceptionally different than Redox GEN-1 cells made using the standard production process. This indicates that the calendering process, with more improvement, has a similar level of critical defects as the standard processing route.

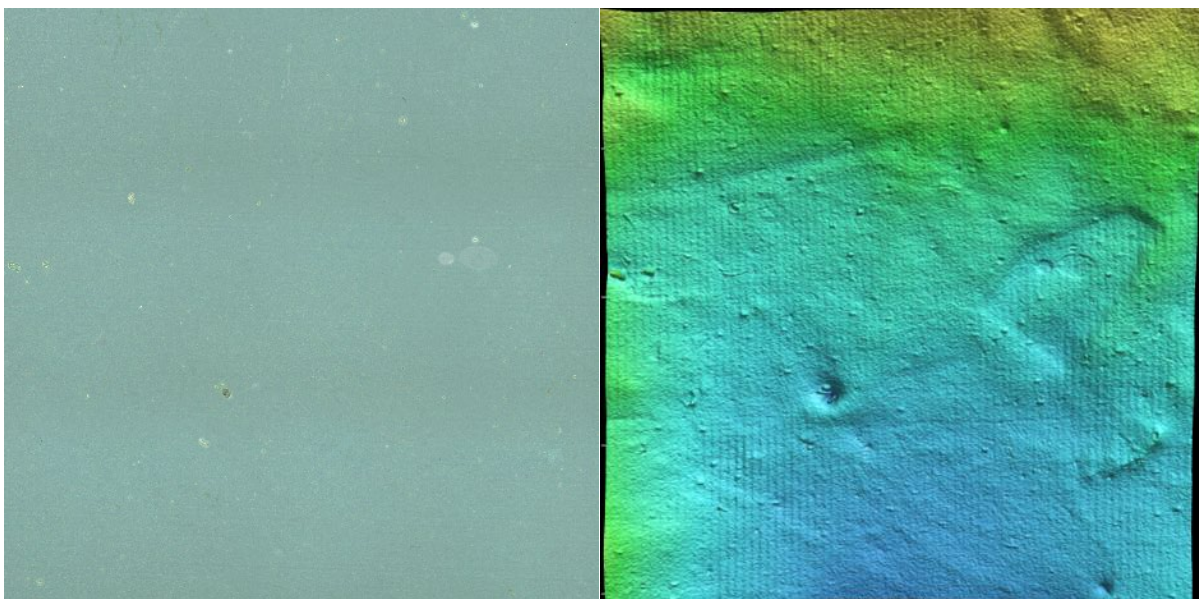


Figure 8. Optical (left) and profilometry (right) images of the electrolyte surface of the medium gap cell used in SOFC testing. The profilometry image is magnified 9kx in the out-of-plane direction to aid in defect detection.

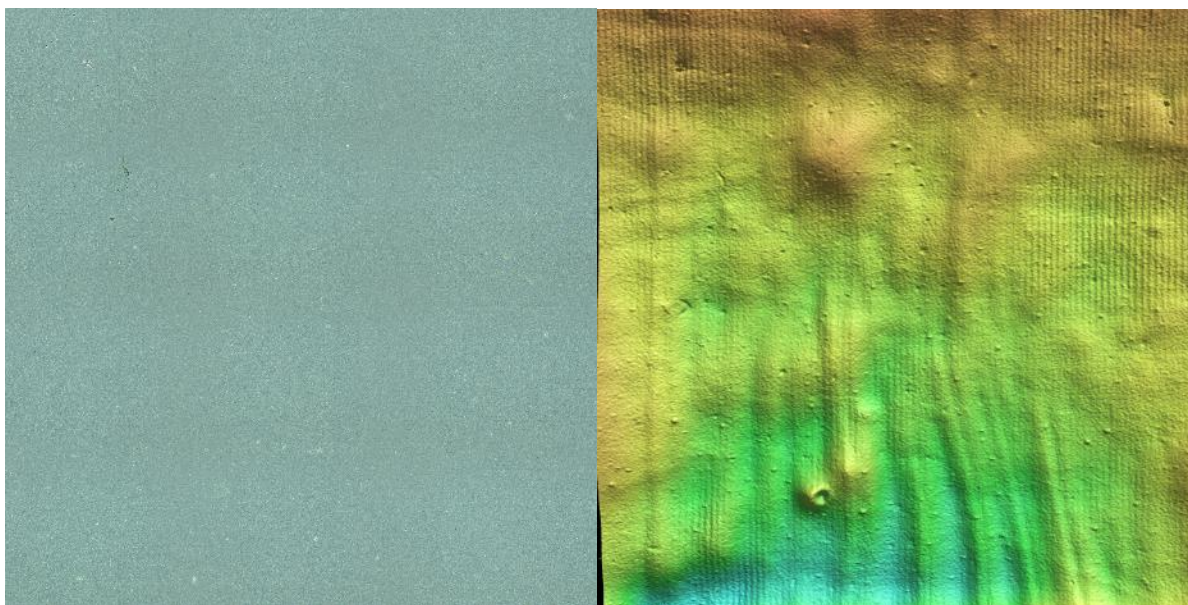


Figure 9. Optical (left) and profilometry (right) images of the electrolyte surface of the small gap cell used in SOFC testing. The profilometry image is magnified 9 kx in the out-of-plane direction to aid in defect detection.

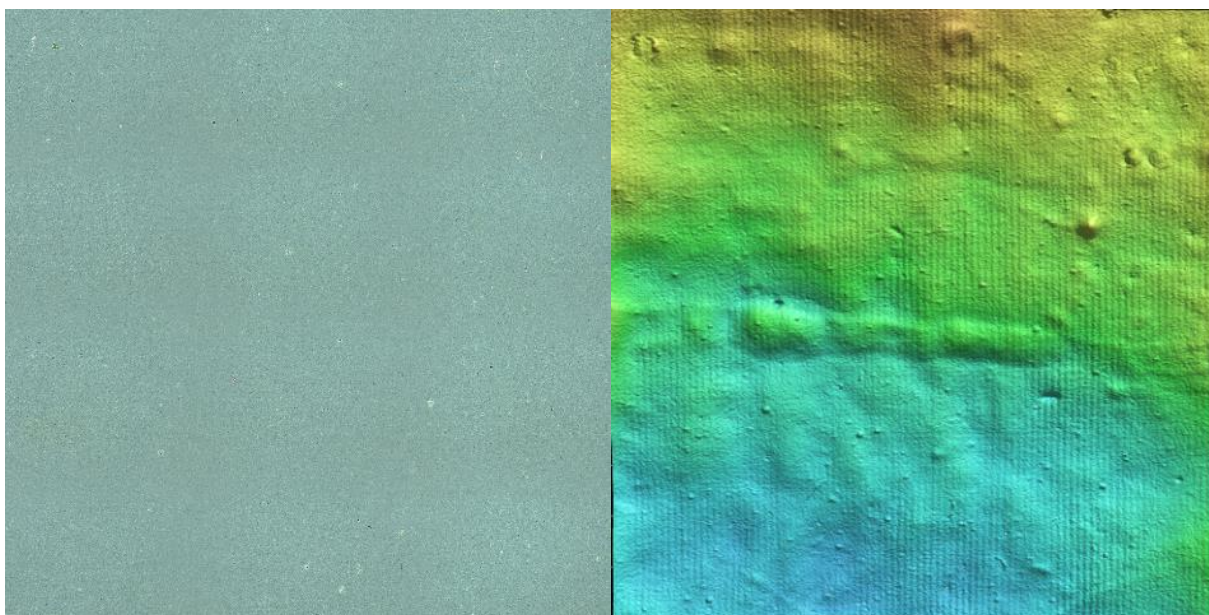
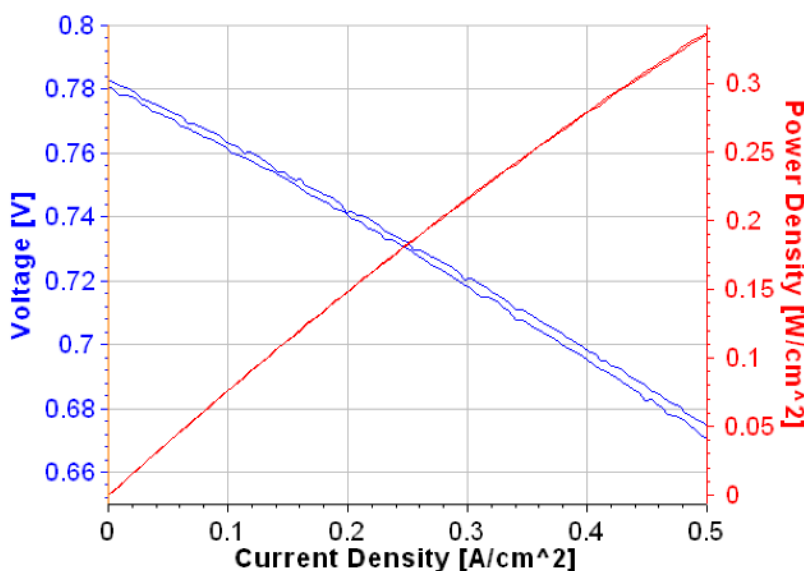


Figure 10. Optical (left) and profilometry (right) images of the electrolyte surface of the large gap cell used in SOFC testing. The profilometry image is magnified 9kx in the out-of-plane direction to aid in defect detection.

A voltage and power density curve for the medium gap SOFC measured at 650 °C after 150 h of operation is shown in Figure 11. The open circuit voltage (OCV), ~0.78 V, is lower than a typical Redox cell, but is, in part, expected for the thinner GDC electrolyte layer used in the

calendering trials. Additionally, defects, such as pinholes or cracks (possibly exacerbated by the horizontal and vertical lines described above), that may be introduced during calendering would further lower OCV. Moving to a thicker electrolyte and improved calendering process, or the slot die process discussed later, are expected to aid in increasing OCV and power density of the SOFC. Despite the lower OCV, the area-specific-resistance (ASR) is comparable to standard Redox cells (described below), and thus the power densities are not significantly lower than in the standard cell.



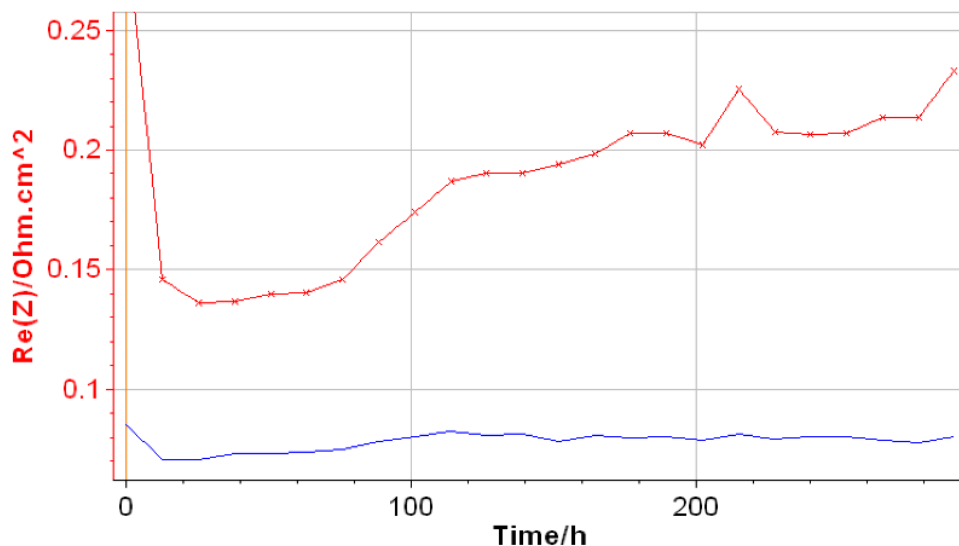


Figure 12. Total (red) and ohmic (blue) ASR contributions to impedance measured at OCV for the “medium gap” SOFC.

Additional ASR curves are shown for the small (Figure 13) and large (Figure 14) gap samples. In these two samples, the ohmic ASR is about $0.11 - 0.13 \Omega\text{-cm}^2$, larger than the medium gap sample. The electrode impedance at 150 h is 0.11 and $0.13 \Omega\text{-cm}^2$ for the small and large gap samples, respectively, which is the same or larger than the medium gap sample. Based on these results, the medium gap sample processing conditions are the best, as they yield the lowest ASR. It is worth noting that all samples showed similar OCV values of 0.78 and 0.785 V for small and large gap samples, respectively. The small gap sample is very similar to the medium gap sample in performance and thus should not be ruled out immediately for further testing in future work.

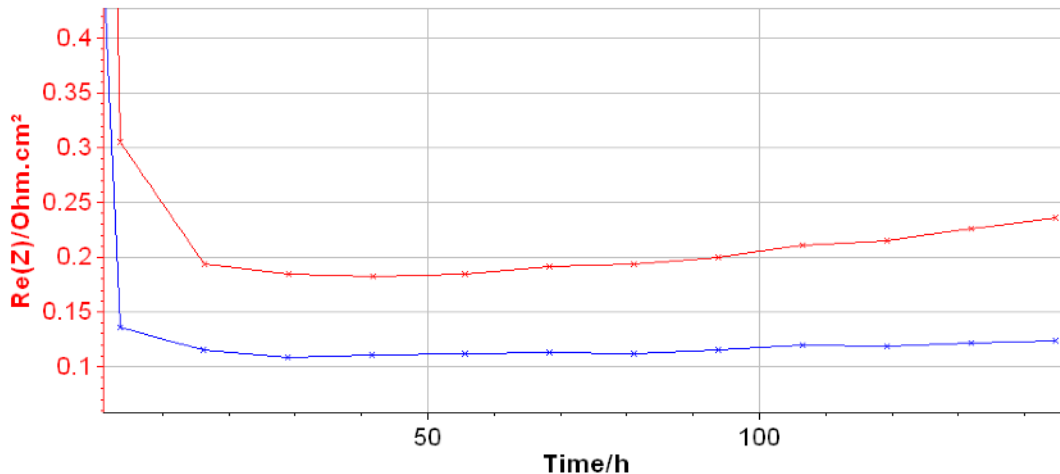


Figure 13. Total (red) and ohmic (blue) ASR contributions to impedance measured at OCV for the “small gap” SOFC.

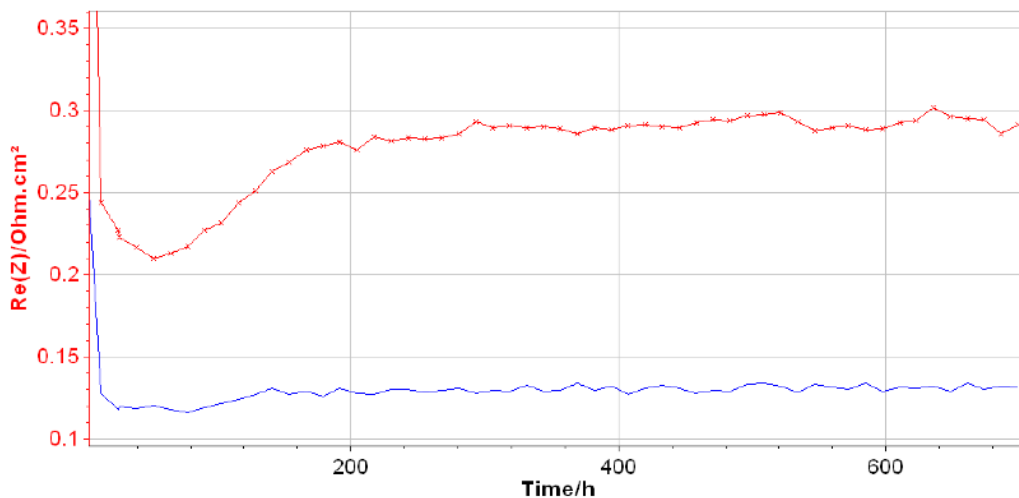


Figure 14. Total (red) and ohmic (blue) ASR contributions to impedance measured at OCV for the “large gap” SOFC.

The long-term performance was also stable for the SOFC measurement up to ~700 h, as shown for the large gap sample in Figure 14. After a burn-in up to about 200 h, the total and ohmic ASR contributions exhibit very little to no measurable change in value. This indicates the calendaring process does not impact the long-term performance of the cells up to the measured conditions shown here. Further work for much longer time frames will be needed to confirm there is not a negative impact of calendaring on long-term performance. The small and medium gap tests were not evaluated beyond ~150-300 h due to limited test stand availability for this evaluation project.

Post-test microstructure images from scanning electron microscopy are shown in Figure 15. Evidence for ASL-AFL poor adhesion during lamination is observed for the medium gap sample, but not in the small or large gap samples. Excellent lamination between electrolyte and AFL layers is observed for all samples in the images. Despite the poor ASL-AFL lamination observed in the image for the medium gap sample, the cell electrochemical performance was the best of the three. Other areas of the sample showed good lamination quality. As described above, a more detailed analysis of adhesion across a larger area of the sample than found in a cursory SEM investigation would be needed. However, the good adhesion observed between layers for the majority of the samples shows excellent promise for this manufacturing process.

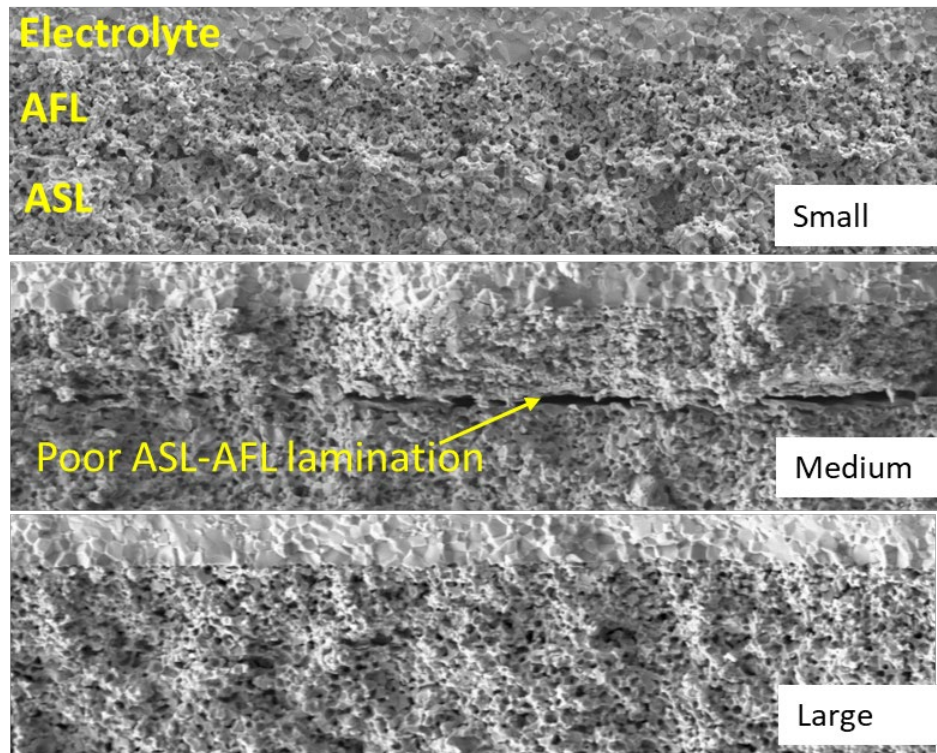


Figure 15. Images of fractured cross-sections of post-tested SOFC cells for the small, medium, and large gap samples.

4.4 ECONOMIC ANALYSIS FOR REDUCED CELL MANUFACTURING COST USING R2R PROCESSING

In this task, we estimated the cost of the R2R processes and compared the results to Redox's standard manufacturing approach to quantify potential costs savings at the cell level. The techno-economic analysis (TEA) methodology conducted for the R2R process was based on design for manufacturing and assembly (DFMA) techniques [B. James et al., Manufacturing Cost Analysis of Stationary Fuel Cell Systems, September 7, 2012]. The DFMA approach models the actual processing procedures used in cell manufacturing, which were modified for R2R processes from extensive TEA modeling work that Redox and Strategic Analysis performed as part of the ARPA-E REBELS program (DE-AR0000494). The process steps considered in manufacturing a cell include slurry preparation, tape casting, lamination and optional slot-die coating, punching (i.e., final green shaping), high temperature sintering of the half cell (i.e., anode and electrolyte), and electrode/contact printing and firing. In the R2R TEA, we considered all of the steps prior to half-cell sintering.

The DFMA process model took into account materials costs, tooling costs, and manufacturing costs. The materials cost associated with SOFC manufacturing accounts for the cell geometry and composition, as well as additives in the slurries, consumables, waste, and yield. The manufacturing cost includes expenses such as the capital cost of equipment used in manufacturing (as well as financing costs), installation cost, maintenance and other expenses, total annual runtime, annual setup time, electrical utility energy cost, process power usage, fully loaded labor cost, and number of employees/line.

A TEA was conducted for two R2R processes and the cell manufacturing cost of each was compared to the standard baseline process (i.e., slurry preparation, tape casting, sheeting, stacking, and lamination with a uniaxial press or isostatic press). The first R2R process we considered was a sequential (in-line) calendering process that laminated the ASL, AFL, and electrolyte layers together in a single, continuous process stream. The second R2R process model was a hybrid calendering and slot-die coating process, wherein the ASL layers are first laminated with calendering equipment and then the AFL and GDC electrolyte layers are subsequently deposited with the slot-die coater.

The cost of equipment per manufacturing line that is needed in the baseline process is similar to that for each of the R2R processes. The stacking process is relatively slow in the baseline process, and therefore as the production volume increases, the number of manufacturing lines needed increases dramatically compared to the R2R processes. The R2R machine speeds used in the modeling are values known to result in acceptable materials as determined from this SBV project's results.

The baseline process and the first R2R process (i.e., calendering) utilized tape cast layers for the ASL, AFL, and electrolyte. The second R2R process (i.e., calendering of the ASL and slot-die coating of the AFL and electrolyte) utilized only ASL tapes. As there is a relatively significant cost associated with tape casting, the second R2R process had an inherent advantage compared to the baseline process and the first R2R process. In fact, at lower production volumes, the cost of the tape cast layers used in the 2nd R2R process was 2/3 that of the baseline process. About half of this extra cost is associated with the materials cost and therefore is still incurred even in the case of the 2nd R2R process.

The TEA model was used to investigate how cell manufacturing costs changed as production volumes varied from ~100,000 cells per year to ~100 million cells per year. This represents a capacity of up to about 8 GW of power. Figure 16 shows the relative cost of each of the modeled R2R processes relative to the baseline process. At the lowest volume of production, the cost of the calendaring process is actually slightly more expensive than the baseline process because of very low utilization for the calendaring equipment relative to the baseline process equipment. The hybrid calendaring/slot-die process, however, is ~12% less expensive than the baseline. This is primarily because there is no AFL or electrolyte tape cast layers in the hybrid process. At higher production volumes, the cost for both R2R processes drops to below 70% of the baseline process. Since the cell cost represents a significant factor in determining the overall power system cost (e.g., as much as ~30% of the total cost), these results demonstrate the true potential for these R2R processes to contribute to an affordable, high efficiency SOFC power system [B. James et al., Manufacturing Cost Analysis of Stationary Fuel Cell Systems, September 7, 2012].

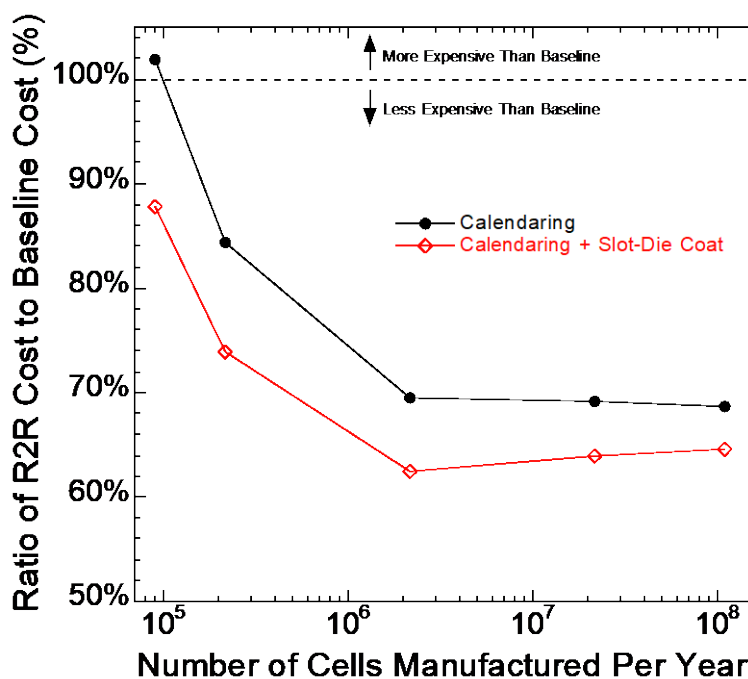


Figure 16. Ratio of the cost of the R2R processes to the cost of the baseline process versus annual production volume.

5. SUBJECT INVENTIONS

One Invention Disclosure (ID#: 201804281), “Roll to roll SOFC Manufacturing for low cost energy generation”, has been filed. This invention discovered and identified processes conditions to R2R laminate cell components into stacks, which demonstrated desirable morphology and comparable electrochemical performance to those from conventional process methods.

6. COMMERCIALIZATION POSSIBILITIES

Redox is working towards commercialization of affordable, efficient SOFC power systems. The R2R processes demonstrated in this SBV project show significant promise as a future route to low cost cell manufacturing. The TEA work has shown that at modest cell production volumes, R2R methods become more cost effective than the standard manufacturing process. Combined with demonstrated electrochemical performance similar to cells made using the standard process, Redox is convinced that R2R manufacturing deserves further development. Future scale-up efforts with ORNL and R2R toll-service providers will further reduce risk and prove out these advanced manufacturing processes.

7. PLANS FOR FUTURE COLLABORATION

ORNL has a number of collaboration plans with Redox to further develop and demonstrate the technology in large format cells. Options include the technology commercialization fund (TCF), Innovation Crossroads, Energy I-Corps, and NTRC Technology Collaboration. The focus will be optimizing processes and generating processing knowledge to streamline SOFC manufacturing and reduce cost in the future to meet DOE's cost targets for a variety of applications.

8. CONCLUSIONS

R2R processes have been developed in fabrication of SOFC electrode components via slot-die coating as well as manufacturing laminates with multiple electrode layers via calendering. While further work is required in optimizing process conditions for best electrode morphology and structure, and cell performance, the success of proof-of-concept demonstrated the viability of manufacturing SOFC via R2R processes, which can significantly increase production throughput and dramatically reduce manufacturing cost. Specifically, we have demonstrated the followings:

- Role of calender set gap examined for three conditions – clear trend in compression percentage observed
- All 4-layer calendered samples resulted in testable SOFCs
- SOFC testing indicated the medium gap set point to be the best based on ASR results, though the small gap sample showed promise.
- Calendered samples exhibited similar ASR values as expected for standard Redox GEN-1 SOFCs and tested for up to ~700 h, showing the calender process yields good SOFC performance
- Defects, such as regions of poor adhesion of layers, observed with optical imaging, electron microscopy, and height profile metrology. Future work with a more automated (not hand-fed as the case here) configuration and additional process condition changes are expected to alleviate these defects.
- A hybrid calendering/slot-die process was shown to be the most cost-effective route to fabrication as compared to only calendering and tape casting, especially at high volumes.