U.S. DEPARTMENT OF ENERGY | Energy Efficiency & Renewable Energy

Advanced Manufacturing Office

High Value Roll-to-Roll Manufacturing

Workshop Summary Report

December 2-3, 2015

Alexandria, VA

The DOE Office of Energy Efficiency and Renewable Energy (EERE)'s Advanced Manufacturing Office partners with industry, small business, universities, and other stakeholders to identify and invest in emerging technologies with the potential to create high-quality domestic manufacturing jobs and enhance the global competitiveness of the United States.

This document was prepared for DOE/EERE's AMO as a collaborative effort between DOE AMO and Energetics Incorporated, Columbia, MD.

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1. Workshop Executive Summary

Executive Summary

The Department of Energy's (DOE) Advanced Manufacturing Office (AMO) held a workshop on High Value Roll-to-Roll (R2R) Manufacturing to gather inputs from stakeholders on the vision of future opportunities and technical challenges facing development and scale-up of new manufacturing processes, sensors, algorithms, materials, and equipment that can make step-change improvements of system performance in R2R manufacturing. Several themes emerged throughout the workshop, all of which are interconnected and contribute toward the vision of rapidly deploying cost-effective, innovative R2R manufacturing technologies capable of increased yields while maintaining stringent quality and cost control.

Key themes include:

- **Process Speed:** Significantly increase the manufacturing throughput of R2R by factors of 5 or even 10 as compared to the current state-of-the-art. This process would be applicable to many clean energy technologies, such as photovoltaic and batteries.
- **Quality Control and Metrology:** Cost-efficient metrology and quality control (QC) technologies that enable R2R processes to manufacture clean energy products with higher yield cost efficiently. New metrology and QC technologies will have high-resolution (micron-scale) measurement capable sensors with high-accuracy detection for faster areal measurements of quality and contamination detection.
- **Multi-layered Material:** A cost-efficient R2R process that is able to produce multilayered, functional, and tailored structure in ceramic/metal/polymer membrane systems for gas/liquid separations with uniform and predictable properties.
- **Transition to R2R:** A cost-efficient approach to transition plate-to-plate processes to R2R, to achieve increased yield while benefiting from cost reduction, with targets such as increase production capacity by a factor of 10 times while reducing manufacturing cost (including energy and labor) by 50%.
- **Continuous Processing:** A cost-efficient R2R process that can fabricate products with multilayer coating at high speeds, with in-line measurement and process control that achieves in-line inspection, monitoring, and tracking at a 100% level.

During the five breakout sessions, a set of priority topics for research and development (R&D) emerged from discussions on the challenges and R&D needs identified for high valued R2R manufacturing, as seen in Figure 1. These topics represent areas where a concerted R&D effort could help overcome major material and technology challenges. The topics are summarized below and described in more detail in Sections 2 and Appendix E of this report.

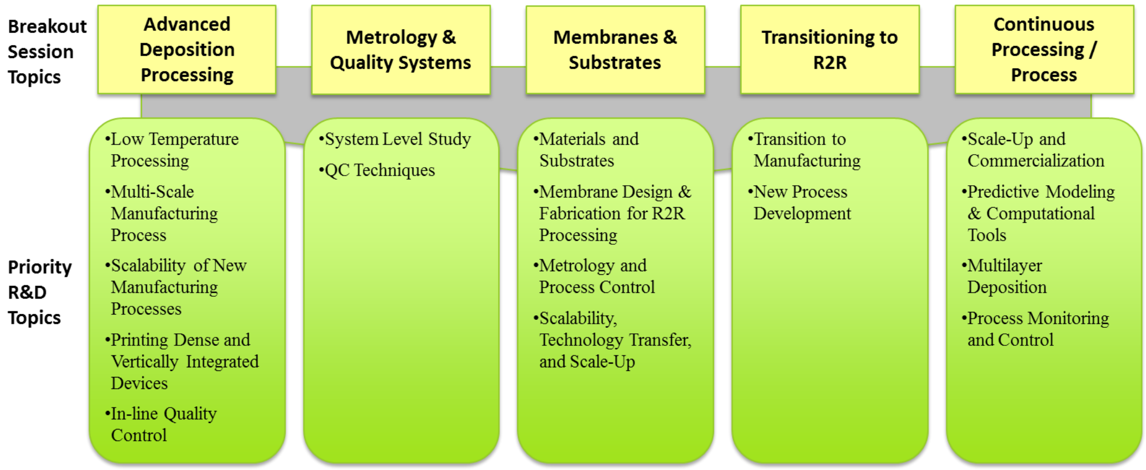

Figure 1. A set of priority R&D topics for high valued R2R manufacturing.

Description of the Opportunity

Roll-to-roll (R2R) manufacturing is a low-cost, high throughput technique for continuous two-dimensional (2-D) deposition of materials over large areas onto moving webs, carriers, or other substrates. Also known as web processing or reel-to-reel processing, R2R creates products on a roll of flexible plastic, glass, ceramic, composite, or metal foil. It can also refer to any process of applying coatings, printing, or other processes starting with a roll of a flexible material and re-reeling after the process to create an output roll. Of interest is to focus on R2R manufacturing advances which would have a significant impact on clean energy technology manufacturing such as thin film flexible solar photovoltaics (PV), battery and capacitor electrodes and electrolytes, organic light emitting diodes (OLEDs), reflective optical films for concentrated solar power, building structures with embedded PV or energy storage, embedded sensors for better energy management, low cost efficient windows, and various membrane applications (e.g., fuel cells, biofuels separations, water desalination, and industrial separations).

The potential impact of R2R manufacturing on just even one clean energy technology platform is significant. DOE estimates that replacing traditional lighting systems with phosphorescent OLED lighting will reduce energy by 0.22 quadrillion British thermal units (quads), saving domestic consumers $20 billion and reducing environmental pollution emissions by 3.7 million metric tons.[1]

The high-value R2R manufacturing topic will focus on technologies and methods to improve yields and reduce costs by developing advanced approaches for deposition and processing, precision patterning processes, ever-smaller and finer size scale such as high-resolution in-line metrology techniques, and embedded thermal energy minimization and enhanced cooling.

Research and Development Needs

Various R2R manufacturing innovation platforms cut across the industrial base and are appropriate for focused development programs. In addition, multi-partner collaborations may be able to address and overcome the key foundational challenges to radically advance R2R and associated industrial manufacturing technologies.

[1] U.S. Department of Energy, SBIR Advances, "New OLED Lighting Systems Shine Bright," Save Energy, 2013.

Experts have suggested that greatly improved R2R process technologies could have cross-cutting applications relevant to the energy industry and beyond. R2R manufacturing has been a major driver for competitive processing at required volumes for a range of commercial products, such as multilayer capacitors and coated Mylar™. R2R is used in paper, electronics (both passive and active), building materials, chemicals processes (membrane separations, petroleum cracking), PV, and textiles.

Manufacturers are seeking tools, sensor technologies, and new R2R processes and equipment to enable higher speed processes, greater width dimensions, and finer resolution for precisely aligned multilayer materials and designs. Manufacturing requirements also call for near-100 percent quality assessment for chemistry, build, process completeness, and performance of manufactured products. New equipment technologies, substrate technologies, take-up methods, and processes, plus advanced in-line and post-process metrology methods are needed to meet these requirements. New technologies must also be integrated with new and existing software tools, sensors, communication, and automation.

Workshop Overview

DOE's AMO held the High Value Roll-to-Roll Workshop on December 2-3, 2015. Representatives from industry, academia, DOE national laboratories, and non-governmental organizations gathered in Alexandria, Virginia to hear keynote addresses, expert panel discussion, and participate in workshop breakout sessions. Discussion topics focused on challenges and opportunities for new technology development that will enable R2R to be a manufacturing differentiator in the high-yield clean energy material and product production.

The AMO within DOE's Office of Energy Efficiency & Renewable Energy (EERE) partners with private and public stakeholders to improve U.S. competitiveness, save energy, create high-quality domestic manufacturing jobs and ensure global leadership in advanced manufacturing and clean energy technologies. AMO invests in cost-shared research, development and demonstration (RD&D) of innovative, next generation manufacturing processes and production technologies that will improve efficiency and reduce emissions, reduce industrial waste, and reduce the manufactured product life-cycle energy consumption. AMO is particularly interested in the challenges associated with advanced manufacturing technology that might be overcome by pre-competitive collaborations conducted via a Manufacturing Innovation Institute (MII).

AMO seeks to gather input from stakeholders on the vision of future opportunities and technical challenges facing development and scale-up of sensors, processes, and equipment that can make step-changes to improve yields while maintaining product quality produced by R2R manufacturing processes. AMO also seeks individual input on challenging performance metrics and identification of key problem sets to be addressed. At this workshop, participants identified mid-Technology Readiness Level (TRL) and Manufacturing Readiness Level (MRL) R&D needs, market challenges, metrics and impacts, and other considerations for high value R2R manufacturing. The intent is to define critical cross-cutting problems/barriers that, if successfully addressed, represent a step change beyond current state-of-the-art.

The specific purposes of the session were to:

- Identify pathways to demonstrate technologies and mitigate risk to accelerate real-world, specific applications.
- Identify proper investments to enable innovations and development activities, moving technologies from TRL/MRL 3-4 to TRL/MRL 7-8 and toward deployment.
- Establish clear goals to demonstrate and transition R2R manufacturing technologies to industry, with clear road maps towards cost competitiveness for high throughput manufacturing.

- Identify new technologies to lead to less energy intensive manufacturing and facilitated scale-up due to reduced factory capital costs.
- Identify pathways to demonstrate manufacturability of new R2R products at pilot-scale, enhancing development of manufacture-able products such as membranes, electronics, composites, etc. which serve to enhance the broad-base cross-cutting industry sector.
- Discuss facilities that can be offered with test-beds for in-the-loop hardware testing in a relevant environment (TRL/MRL 5) to enable functional demonstration of high throughput capabilities for cross-cutting applications. These facilities will have industry-wide impacts.
- Identify new technology advancements, such as precise depositions/patterning designed and developed at multiple scales.
- Discuss efforts that will potentially produce prototype metrology platforms that can be developed and demonstrated in-line.
- Introduce manufacturing communities that support materials and systems development for R2R manufacturing to the U.S. DOE AMO and EERE offices.
- Strategize how best to leverage R&D among the U.S. DOE and other Federal agencies.
- Encourage discussion and networking among leaders in the field.

Panel Discussions

A panel of subject matter experts (SME) provided their insights on the capability needs and research trends on R2R manufacturing. The panel on the first day composed of experts from both non-government organizations (NGO) and industry. Biographies of the panelists can be found in Appendix D.

Highlights of the SME panel include:

- Dr. Fabio Albano, Director of Research & Development at Xalt Energy and Energy Power Systems, emphasized the possible impact of R2R processes on lithium-ion battery electrode manufacturing. R2R is potentially a new and low cost manufacturing alternative. If successful, this will help the electric vehicle (EV) market achieve key goals such as EV fuel efficiency of 34.1 miles per gallon (MPG) by 2017 and 50.1 MPG by 2025 at a battery cost point below $250/kiloWatt hour (kWh). The availability of lithium-ion batteries will increase the fraction of renewable energy being implemented in grid applications, as grid application of renewable energy often requires an energy storage component. Dr. Albano concluded his remark by challenging the group to keep their eye on the $80 billion battery market opportunity, of which advanced R2R manufacturing will give the U.S. a global competitive edge[2].
- Dr. Paul D. Fleming, Professor of Flexible Electronics Applications & Technology (FEAT) Center at Western Michigan University, spoke about the role FEAT plays in the Manufacturing Innovation Institute for Flexible Hybrid Electronics. Revolving around the theme of advancing R2R technologies, Dr. Fleming discussed FEAT's three focus areas: educate the future printed electronics (PE) workforce, research in PE and Flexible Hybrid Electronics (FHE), and create an environment to foster innovation for start-up companies.
- Dr. Venkat Srinivasan, Staff Scientist at Lawrence Berkeley National Laboratory, focused his discussion on the role of high-performance computational simulation and how it will facilitate the identification and application of new materials into R2R processes. Computation simulation will make new discovery of materials possible, while taking into account the complexities and requirements of device fabrication. This will ensure that new material performance will meet device performance but also accommodate R2R process conditions.

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[2] C. Pillot, "Battery Market Development for Consumer Electronics, Automotive, and Industrial: Materials Requirements and Trends," Avicenne Energy Report, 2013.

- Dr. Brian B. Berland, Chief Science Officer at ITN Energy Systems, begun his discussion with a review of the different R2R process applications. For example, manufacturers can use R2R to fabricate separation membranes for water purification and flow batteries. He emphasized the potential of R2R processes to offer the advantage of a smaller physical footprint and enables product integration versatility. Dr. Berland concluded his remarks with a review of some R2R challenges across the technology space. For example, additional research is needed to develop intelligent process controls that would provide the required high throughput and yield to realize economies of scale. Also, new technologies are required to develop nano-dimensional material control for performance optimization.
- Dr. David Wood III, Senior Staff Scientist at Oak Ridge National Laboratory (ORNL), reviewed on-going efforts at ORNL to address R2R processing issues relevant to industry. One example is on-line and off-line non-destructive evaluation (NDE) for coating quality control (QC) improvement and long-term performance enhancement. ORNL is also investing in approaches to optimize web speeds for different deposition methods and applications. He also shared, from his perspectives, areas of R2R where additional research and investment is required. This includes technology that can identify locations in R2R processes where in-line QC is needed. Also, an approach is needed to increase the success rate of technology transfer of developed in-line non-destructive evaluation (NDE) and QC methods to industry.
- Mr. David Ventola, Director of Business Development at Babcock & Wilcox MEGTEC, focused his discussion on the opportunities and challenges of battery electrode manufacturing via R2R. While R2R has the benefits of continuous operation, the challenge will be the shift from batch to continuous process while minimizing changeovers. The transition of process models for scale-up from the laboratory environments to production is also a challenge. The drive towards higher efficiencies and yields will push toward wider webs and faster line speeds.

Workshop Discussions and Breakout Sessions

The workshop discussions provided AMO with further information on both cross-cutting and specific technology R&D challenges. Additional discussions on basic rationale for an MII, consistent with the missions of the DOE, also took place at the workshop. Federal presentations given at the workshop are available at http://energy.gov/eere/amo/downloads/workshop-high-value-roll-roll-hv-r2r-manufacturing-innovation-december-2-3-2015. Five breakout sessions were conducted, as shown below.

- **Advanced Deposition Processes:** High throughput equipment with formats applicable to nano/atomic layer through thin-film to thick-film, i.e., nano through macro size-scales. Development would focus on tools to feed precursor solutions and slurries at sufficient rates while controlling the rheology of these materials; simulation and design tools; control(s) for feedback and adjustment; materials drying, curing, and heat treating accessories; incorporation of concurrent/simultaneous processes using additive and subtractive techniques; development of atmospheric and vacuum processing; precision alignment and registration; and lithographic imaging and etch/deposition.
- **Metrology and Quality Systems:** Commercial enterprises, which incorporate R2R manufacturing processes must detect, control, and otherwise eliminate potential quality issues within products. Technology development to enable higher resolution and increased data capture/processing rates is needed to enhance inspection for mechanical defects such as pinholes and cracks, measurement of electrical properties such as resistance measurement, and inspection for inter-layer delamination and voids. All data would be integrated into process control and feedback systems; technologies would help correlate defects to performance.

- **Continuous Processing:** R2R additive manufacturing such as what is used for electronics applications in the process of larger format flexible displays, detectors, and stretchable/conformable sensors, etc., as well as other composites supportive of other industrial sectors as for buildings, chemical process and a wide range of other applications and products. This technology will lead to a fundamental change in the manufacturing of these systems from plate-to-plate standard lithography or other batch processes to continuous R2R processing.
- **Membranes, Substrates and Associated Functional Materials and Cross-cutting Applications:** Rigid and flexible, thick-firm and thin-film substrates and deposited functional materials, such as glass, Al_2O_3, AlN, Si_3N_4, SiC, GaN, MgO, ZrO, metal foils, polymers, membranes, catalysts, photovoltaics, etc. have cross-cutting applications for nearly all energy technologies. Applications include photovoltaics, electrodes and electrolytes, smart/mesoporous membranes, nano-scale separators, sensors, piezo-electrics, fuel cells, flexible displays, gas (H_2, CO_2, etc.)/liquid/low-energy/chemical catalyst filled separation/chemical process membranes, other composite systems and ultrathin membranes, or other nanotechnologies tailored for new materials used for energy applications. All could be produced using high-value R2R methods. Additionally, improved R2R processes are needed to address substrate properties such as tensile strengths, surface finish and imperfections and release character, impurities, inconsistencies in substrate thickness, flatness, and planarity of webs/membranes/substrates.
- **Process and Equipment Needs for Transitioning Plate-to-Plate to R2R:** Technologies and products supportive of energy storage and generation, chemical separation, flexible circuitry for displays and sensors, and many others can be fabricated using a mixture of R2R and batch plate-to-plate processes. Associated processes may involve (such as for microelectronics) lithography printing, chemical etching and etch-back, doping, trimming/properties adjustment, and other processes. Equipment development to enable a complete transition to continuous R2R process approaches would dramatically advance the technology. However, previously this has not been successful, such as has been the case for flexible digital detectors and for many display applications based on a silicon thin-filmed transistor arrays. Development of new equipment, processes, technologies, and supportive hardware/software/accessories, such as; motor controls for web speed, tensioning, material take-up, etc., motors and drive systems, punching and other post formatting tools, etc. are required for this transition to be successful.

Participants in each breakout session answered a different set of questions that were appropriate for the topic. Summaries of the breakout group discussions and questions posed are outlined in the following chapters. The Appendices include the meeting Agenda (Appendix A), a combined list of participants from all the breakout groups (Appendix B), and an acronym list (Appendix C).

Collaboration/Partnerships

The participants agreed that an institute or a National Laboratory led Private-Public collaborative consortium is required to properly investigate high value R2R manufacturing, as a single entity cannot afford to undertake such a large effort individually. Further, breakout group participants identified the need for an environment where shared resources such as supercomputing capability and SMEs are accessible by industry and researchers. Multiple partners: would be involved, including industry, government, academia, non-profit, and national laboratories. The result of this collaboration is an environment that supports full-scale demonstration of new R2R technologies to facilitate commercialization.

Possible Overarching Metrics Resulting from Collaborative Efforts

Example technical goals for R2R technologies include demonstrating microelectronics manufacturing throughput in excess of 100,000 mm^2/min using concurrent dual-side deposition/processing without the need for web reroll/rework and demonstrating a greater than 5%/yr. reduction of material costs through product design and fabrication of smaller scale devices, kerfless cutting techniques, and quality enhancements. Potential metrics to measure technology development by the consortium would include:

- Enhancement of resolution capability to enable registration and alignment of multiple layers of coatings/patterns and printing of features and device architectures on these layers at five micrometers within 5years on a path to one micrometer within 10 years, which are scalable to commercial products.

- Develop scalable R2R processes for solution deposition of ultra-thin (<10 nanometers) dense films and to fabricate discrete multilayer composites of different materials and dimensions using concurrent or series processing.

- Development of tools and processes to increase throughput, such that over the next 5 years, an increase in the throughput of R2R processes by 5 times for batteries/capacitors and 10 times for printed electronics.
- Decrease cost of R2R manufacturing by 50% over 5 years, including energy and labor cost. Examples include: for PV, reduce the cost of PV in the next 5 years to achieve $0.5/watt; for desalination applications, the cost is envisioned to be $0.25/cubic meter; for battery applications, the cost target for proton conducting membranes is less than $50/square meter; for processing OLED lighting panels, products at $10/kilolumen and greater than two times cost reductions in with less than 10-6 g/m2 deposition per 24 hours.

- Establish Quality Control methodologies, which for chemical based R2R process, increase QC resolution to <10 meter at 100 ft/min in the next 3 years and <1 meter at 300 ft/min after 5 years and for vacuum deposition processes, increase QC resolution to 100 nm at 2 ft/min in the next 3 years, and 10 nm at 20 ft/min after 5 years. Further these methods will enable property prediction in-process.

- Overall enhanced Product Performance. As example, for chemical membranes, in the next 10 years, increase durability by 10 times, selectivity up to 5 times, and permanence by 10 times compared to current state-of-the-art processes.

2. Summary of Results

Advanced Deposition Processing and Printing for ALD, Thin, Mid and Thick-film Size-Scale

This breakout group focused on four key topic areas related to *Advanced Deposition Processing and Printing for ALD, Thin, Mid and Thick-film Size-Scale*:

- Topic 1: Vision and Goals
- Topic 2: Barriers and Challenges
- Topic 3: Research and Development Needs
- Topic 4: Priority R&D Topics

For each area, one focus question and several additional questions were posed. Individual participant's views and responses were captured using a compression planning and brainstorming process. Highlights of discussions are outlined below; the full results of discussions are provided in the Appendix E.

Several themes emerged throughout the session, all of which are interconnected and contribute toward the vision of a cost-efficient R2R process that has the capability to significantly increase the manufacturing throughput by a factor of five or even 10 as compared to the current state-of-the-art. This process would be applicable to many clean energy technology, such as photovoltaics and batteries.

Vision and Goals

On the discussion of vision and goals, key recurring themes are highlighted below. The full results of the discussion are provided in Table A-1 of Appendix E.

- **Manufacturing Throughput/Yield**
  - **Speed Increase:** Complete R2R coating at five-times faster speed for batteries/capacitors and 10 times faster speeds (many orders of magnitude) for printed electronics (atomistic size). In order to enable increases R2R coating speed, researchers need to understand the limitations of materials coating – what needs to change in the materials to change the speed?
  - **Increase in Yield and Uniformity:** For supercapacitors and semiconductors, one goal is to increase both manufacturing yield and QC. Uniformity is tied to manufacturing yield and the vision is to increase both by at least an order of magnitude.
  - **Printing Solar R2R:** One proposed goal is to beat the current DOE goal of $1/watt for solar printed R2R, by aiming for $0.5/watt in five years. This would require advancement of both the coating and the solar technologies. An interesting proposal was to transition newspaper printing mills to be able to print solar cells.
  - **Other Performance Metrics:** Besides speed and cost goals, other performance metrics can be set for R2R. These might include the amount of energy used per product produced, or the cost to manufacture the product to establish a baseline. Developing a cost to performance ratio metric will help to promote the products to industrial users. Also, it may be possible to model or simulate specific metrics.
- **Reduce Manufacturing Cost**
  - **Lower/Same Cost ($/m^2 goal):** The speed increase noted above should not increase cost above feasible levels. For example, the battery industry is limited in speed because of the requirements in the drying process – it would be longer and more expensive. Setting a

$\$/m^2$ goal along with the increased speed goal or other performance metrics would help ensure cost is not prohibitive.

- **New Manufacturing Technologies and Processes**
  - **Flexibility:** Flexibility is a main goal for both the materials and products for R2R. It is very challenging to deposit high quality materials on flexible substrates. Advanced ALD will be needed to use high quality substrates and not affect quality. For future electronics, smaller and more flexible features will be required. Finally, scalability and flexibility have to be addressed up front; the concept must be scalable.
  - **Improved Process and Material Quality:** Multiple visions and goals were related to improving the processes and materials used for R2R. These include developing low cost advanced deposition processes with very high quality substrates, achieving interconnected multi-layers in printing, and achieving multi-layer pattern coating on thin substrates. Another goal would be able to go from cheap precursors to manufactured thick film in one R2R step, with tuned composition. Additionally, a vision is to develop a mixing technology for slurry-based deposition where the mixes are uniform, thus ensuring mixture purity.
  - **ALD R2R for Batteries:** One possible use for atomic layer deposition (ALD) R2R processes would be integration into battery production lines, where they would be coating highly porous substrates. However, many technical barriers would have to be addressed.
  - **One Micron Scale and Below Printing:** Over the next five to 10 years, it will be needed to enable printing features and device multilayer-alligned architectures at a one micron scale. For future electronics, below one micron applications (e.g., imprinting, patterning of dielectrics) at the 100 nanometers range will allow for multilayer and more sophisticated materials. It was suggested that the technology to go to the 50 nanometers range currently exists at low technology research level (TRL) ; it just has to be scaled.
  - **Wearable Technologies:** This is a focus area of the recently established DoD Flex-Hybrid-Electronics (FHE) Manufacturing Innovation Institute (MII), known as NextFlex[3]. More investigation would be needed regarding wearable technologies in film or textiles, but this represents a logical expansion area for R2R. These type applications include monitoring technologies, typically integrated into a garment to monitor the wearer's heartbeat. More information would be needed to determine if R2R could play a role in this area.
  - **No Dry Room Needed:** Ideally, materials will be able to be processed outside the dry room without all the current environmental constraints.
  - **Better Composition Detection:** One goal is to be able to detect structure and/or chemical composition at high throughput, especially in a three dimensional (3-D) format.

Challenges and Barriers

On the discussion of challenges and barriers, key recurring themes are highlighted below. The full results of the discussion are provided in Table A-2 of Appendix E.

- **Deposition Processes/Equipment:** There is a lack of models that effectively represent the deposition process to understand the impact of introducing alternative materials/process routes in range of environments (e.g., humid, dry, hot, cold). Also, printing at the nanoscale linewidth and thickness (under room temperature and pressure conditions) is a challenge. An approach is required to adapt R2R for "small" batch operations so that the operation is more modular. Finally, new processes are needed for dry manufacturing and deposition of materials on a R2R platform, where the control of composition, thickness, and cost are well understood. Further, there is a lack

[3] NextFlex, http://www.nextflex.us, Information Sheet, November 2015

of new materials and processes for low temperatures (<200º C) processes which are needed for applications such as solar cells.

- **Scalability/Flexibility of New Manufacturing Processes:** There is a lack of established infrastructure and process that facilitates the demonstration of new manufacturing method scalability; for example, demonstrating a new R2R ALD process at practical line speeds.
- **Materials/Substrate:** New materials and substrates are needed for ALD, as driven by manufacturing processes and application requirements. For example, new materials are needed that can dry fast without cracking and a new ability is needed to grow near single crystal quality films on R2R substrates. QC is also an issue, as slight changes in raw materials can cause large changes in products.
- **Fundamental Chemistry/Properties:** An approach to assess long-term/life-cycle properties of materials and systems when working at the R&D/laboratory level is required. A lack of understanding of the process and limitations at the fundamental level and origin of stochastic variability is also barrier.
- **Defect Identification/Inline Control/Monitoring:** A new real-time capability is needed to control in-line quality R2R process parameters. Developing an approach to register/align multiple layers of coatings/patterns in a reproducible way, for a wide range of features (nanometers to millimeters) is also a challenge. This will also require new technology for metrology.
- **Printing:** One barrier in this area is the inability to print a vertically interconnected multi-layered device below one-micron scale.
- **Other Challenges and Barrier:** There is a lack of collaboration in this highly competitive environment. Accessible demonstration facilities containing state-of-the-art and cutting edge tools are needed to enable companies to assess new technologies without the upfront risk.
- **Cost Factors:** High cost of investigating new materials and R2R processes is preventing exploration. Cost is also a barrier that limits access to tools and expertise.

R&D Needs

On the discussion of R&D needs, key recurring themes are highlighted below. The full results of the discussion are provided in Table A-3 of Appendix E.

- **Cross-Cutting R&D:** New methods for "dry" deposition of sub-micron to millimeter scale thick films were identified as an area of interest. These new methods will enable an ability to control porosity and uniformities. Technologies that enable high-speed electrical, optical, and physical property in-line measurement for R2R were also identified as a R&D need.
- **Printing Vertically Integrated Multi-Layer Device Less than One Micron:** Additional investigation is needed to identify the technologies and processes that support thin and smooth ink formulation. Also, additional analysis is needed on interphase composition, integrity, and durability of a vertically integrated multi-layer device. An improved understanding of the relationship between processing parameters with the structure and performance of final layer is required.
- **Low Temperature Processes/Materials:** Further investigation is needed on processes that do not require special environments and are environmentally friendly. Photonic annealing is also an area of interest, and additional insight to its fundamentals is needed. Another area of investigation is ALD precursors that enable low temperature deposition (namely metals and complex oxides).
- **Scalability of New Manufacturing Methods:** A R&D topic of interest is approaches and technologies that enable pathways for inter-connects (printing and imprinting). New scalable methods for compositionally controlled synthesis are also worthy of investigation. There is a continued interest to investigate different ways of optimizing the R2R process, so that yield is

maximized, while scale-up time and cost are minimized. Scalable processes for micron scale R2R patterning of functional materials also require investigation.

- **Effectively Modeling of Processes:** An area of research is to model processing/R2R manufacturing via materials-genome approach.
- **In-Line QC Parameters:** Investigation is needed on small-scale pilot line(s) with in-line metrology for developing improved coating uniformity and/or sub-micron features. Further, in-line QC methods will need to be developed at a small scale, so that it can be evaluated on commercial machines in-situ for feedback and refinement. Finally, additional efforts are needed to identify new methods for high speed (greater than 1,000 meters per minute) sensing of films and fluids, so that defects can be detected.

Priority R&D Topics

A set of priority topics for R&D emerged from discussions on the challenges and R&D needs identified for *Advanced Deposition Processing and Printing*. These topics represent areas where a concerted effort in R&D could help overcome major material and manufacturing process and technology challenges. For each priority R&D topic, the breakout session participants focused their discussion on:

- Key challenges,
- Desired outcomes,
- Appropriate near-term and the long-term R&D steps,
- Performance goals and targets,
- Potential participants and roles, and
- Impact on clean energy industry.

The topics are summarized below and described in more detail in Figures A-1 through A-5.

- **<u>Low Temperature Processing (< 200º C) for R2R of Functional Materials on Polymer Substrates</u>**: The routes for R2R normally require high temperatures, thus requiring more energy and impeding speed increase. This targeted R&D would lower the temperature for the vapor deposition processes of functional materials on polymer substrates, using R2R platforms while maintaining material properties.
- **<u>Multi-Scale Manufacturing Process (using R2R) for a Structural Energy Storing PV/Battery System for a BIPV</u>**: This priority R&D topic involves proving the feasibility of a building envelope that can store enough energy using a R2R produced PV battery system. Although it would be a challenging product, the goals would be to show that a R2R produced structural battery/PV could be produced more cheaply than the conventional version and that energy storing PVs can contribute to DOE’s cost objectives related to grid parity. The initial stage of the collaborative effort includes full scale demonstration to show its disruptive potential. The possibility of military participation is anticipated during the early stages.
- **<u>Scalability of New Manufacturing Processes</u>**: One recurring topic throughout the group’s discussion was that it is necessary for any new manufacturing process to be scalable, otherwise it is not usable. Scalable processes for multiple areas were noted as desired, such as compositionally controlled synthesis or micron-scale R2R patterning of functional materials. The R&D would involve conducting a research scale vs. pilot scale system study, and connecting small scale users with commercial volume users for early vetting and practical scale-up development.
- **<u>Printing Dense and Vertically Integrated Devices</u>**: Targeted R&D in this type of printing would result in vertically integrated devices at the sub-micron scale produced via R2R, while also improving patterning and efficiency. Key challenges include developing the proper scale alignment, registration, patterning, and printing processes, ensuring vias and interlayer

connectivity, and being able to process materials at ambient conditions. Collaborative efforts will include open access user facilities for emerging technologies and task specific industry/academia/national laboratory consortia and partnerships.

- **<u>In-Line QC</u>**: Development of in-line QC would increase productivity, output, and overall product and material quality. The key challenges to the R&D include attaining uniform thickness and the existence of point defects. Outcomes of the research would use sensing that can assess and map 100% of the material, with feedback control and only periodic instead of random defects.

Metrology and Quality Systems

This breakout group focused on four key topic areas related to the use of *Unique Metrology and Quality Systems for Specific R2R Applications*:

- Topic 1: Vision and Goals
- Topic 2: Barriers and Challenges
- Topic 3: Research and Development Needs
- Topic 4: Priority R&D Topics

For each area, one focus question and several additional questions were posed. Individual participant's views and responses were captured using a compression planning and brainstorming process. Highlights of discussions are outlined below; the full results of discussions are provided in the Appendix E.

Several themes emerged throughout the session, all of which are interconnected and contribute toward the vision of a cost-efficient metrology and QC technologies that enable R2R processes to cost efficiently manufacture clean energy products with higher yield. New metrology and QC technologies will have high-resolution (micron-scale) measurement capable sensor with high-accuracy detection for faster areal measurements of quality and contamination detection. While the focus is on new QC and metrology that will be applicable to many clean energy technologies, such as photovoltaics and batteries, the challenges in technology commercialization and scale-up remain.

Vision and Goals

On the discussion of vision and goals, key recurring themes are highlighted below. The full results of the discussion are provided in Table B-1 of Appendix E.

- **Cross-cutting Applications:** One main goal is to adapt existing QC technologies to new material processing, with a focus on clean energy applications.
- **Metrics**: Ideally, there will be metrics that quantify the benefits (e.g., performance, cost, return on investment (ROI), etc.) of new metrology and QC technologies. Examples include metrics that measure the ROI when R2R is used to manufacture hydrogen fuel cell membranes or metrics that quantify the benefits when big data analytics are used in QC and metrology.
- **Measurement Needs and Technologies:** Ideally, t the measurement frequency of rheology would be increased without significant cost increase. A measurement goal is 10 nanometer in-line profilometry in 100,000mm^2 per minute production.
- **Quality Control and Quality Assurance (QA):** A vision of the group is to have a mature physics-based model that accelerates testing for QC and QA. This modeling will integrate feedback and data measured *in situ* on the R2R line.
- **Manufacturing Process and Control:** One main goal is to develop a new control mechanism that includes corrective action feedback loops to fix defects on the R2R line. While these control mechanisms will need to be tailored to specific applications, they will optimize the manufacturing throughput, at both the modular and plant level.
- **Analysis and Simulation:** The vision in this area revolves around improved predictive analytics, where collected data are used efficiently and effectively. This will yield new automation and plant control software that takes a holistic view of the manufacturing plant. This software will not only be integrated into each process, but also the users will be able to simulate different manufacturing operations and modalities.

Challenges and Barriers

On the discussion of challenges and barriers, key recurring themes are highlighted below. The full results of the discussion are provided in Table B-2 of Appendix E.

- **Scale-up:** Technology scale-up is a key barrier. Insufficient understanding and familiarity of the manufacturing process is preventing the translation and application of laboratory measurements to in-line production. Also, insufficient understanding and familiarity prevents assessment of the impact of process inputs on process energy use, quality, and throughput of the manufactured product. Also, the lack market size, which implies insufficient production volume, will inhibit scale-up as firms cannot justify the capital cost required to upgrade original equipment with automated QC/QA.
- **Metrics and Standards:** Standard operating procedure (SOP) for metrology and quality systems for R2R does not yet exist.
- **Measurement Technology:** Currently, the technology that provides non-visual imaging/areal measurements in a R2R process is immature, too slow, or not amenable to the R2R process environment.
- **Data Analytics:** Current analytical approaches cannot handle the data volume. Companies are reluctant to invest in data analytics, as the ROI is not clear.
- **Material Science:** One challenge is an insufficient understanding of device physics. Better theoretical and empirical understanding of material behavior is required.
- **Material and Process Connection:** The capability of today's metrology sensor and techniques are inadequate. For the known defects, metrology techniques are not available. For the unknown defects, additional insights are needed for if or when metrology techniques would apply. Also, predictive analytics that correlates material behavior to R2R processes to product performance does not exist.

R&D Needs

On the discussion of R&D needs, key recurring themes are highlighted below. The full results of the discussion are provided in Table B-3 of Appendix E.

- **Materials and Substrates:** Areas of investigation in materials and substrates include functionalization methods for polymer substrates, additive R2R processes for novel substrates (e.g., ceramics, glass, advanced polymers), and high-speed, low-cost R2R processes for membranes for batteries and filter applications. Furthermore, printable materials with designable properties (e.g., optical, mechanical, electrical, thermal, and chemical) are also of interest.
- **Membrane Fabrication:** New fabrication R2R methods need investigation to enable membrane production with high selectivity, specifically for fuel cell or CO_2 capture applications. Additional modeling will also be needed to engineer new polymers. Furthermore, research is required in technologies and processes for high-throughput, rapid (<1 second) in-line characterization of structure/morphology of buried interfaces.
- **Metrology and Process Control:** In the area of metrology and process control, a more scientific understanding of different R2R processes (e.g., deposition, drying, sintering, etc.) are needed. Also, additional investigation is required to lead to new processes for measuring membrane performance indirectly during manufacturing. Over all, research is needed to identify technologies that enable in situ and large-area characterization of all parameters, including molecular packing, phase segregation, pore distribution size, and conductivity.
- **Scalability:** In order to facilitate the scale-up of new technologies from laboratory, additional research will be required on new models to better understand how to use nanoscale materials

processing to meet performance goals and materials growth pathways. An objective will be to reduce the R&D cycle time by 50%.

Priority R&D Topics

A set of priority topics for R&D emerged from discussions on the challenges and R&D needs identified for *Unique Metrology and Quality Systems for Specific R2R Applications*. These topics represent areas where a concerted effort in R&D could help overcome major sensor technology, algorithm, and process engineering/science challenges. For each priority R&D topic, the breakout session participants focused their discussion on:

- Key challenges,
- Desired outcomes,
- Appropriate near-term and the long-term R&D steps,
- Performance goals and targets,
- Potential participants and roles, and
- Impact on the clean energy industry.

The topics are summarized below and described in more detail in Figures B-1 through B-2.

- **System Level Study:** This R&D topic focuses on solving the challenge of real-time in-line defect detection. This will require system integration of process science, materials science and metrology. When complete, not only the line performance will be optimized, but also the yield will increase while maintaining minimum device defects.
- **QC Techniques.** This R&D topic will delineate the QC needs for R2R materials, with a focus on areal and linear point measurement technologies that can make detections at line speed. When completed, this will produce Six-Sigma QC for R2R process for clean energy materials.

Membranes and Substrates and Associated Functional Materials and Cross-cutting Applications

This breakout group focused on four key topic areas related to *Membranes and Substrates and Associated Functional Materials and Cross-cutting Applications*:

- Topic 1: Vision and Goals
- Topic 2: Challenges/Barriers
- Topic 3: Research and Development Needs
- Topic 4: PriorityR&D Topics

For each area, one focus question and several additional questions were posed. Individual participant's views and responses were captured using a compression planning and brainstorming process. Highlights of discussions are outlined below; the full results of discussions are provided in the Appendix E.

Several themes emerged throughout the session, all of which are interconnected and contribute toward the vision of a cost-efficient R2R process that is able to produce a multilayered, functional, and tailored structure in ceramic/metal/polymer membrane systems for gas/liquid separations with uniform and predictable properties. Low cost and high performance is key. For example, one objective is to produce low cost, very thin R2R substrates (about ~1 millimeter thick) with sufficient mechanical integrity for ease of handling (without wrinkles or breaks) and moderate temperature processing (150-300°C).

Vision and Goals

On the discussion of vision and goals, key recurring themes are highlighted below. The full results of the discussion are provided in Table C-1 of Appendix E.

- **Membrane Design:** One vision is membranes that are composed of a multilayer polymer, with various additives for improved performance and durability, tailored to applications such as fuel cells, batteries, flow batteries, etc. These membranes must also withstand harsh and challenging operating/storage environments. Lastly, these membranes must be defect-free, thin (micron scale), and cost-effective (at a component level).
- **Substrate Development:** One goal is to develop substrates that have a low-coefficient of thermal expansion and can operate at high-temperature regimes. Another goal is to develop substrates that when produced via nanoparticle/phase alignment by external field can achieve enhanced through-thickness properties such as electrical conduction, thermal conductivity, ionic conduction, and high dielectric properties. Ideally, direct coating of smart materials on porous substrates with multilayers (e.g., in-line, controlled penetration) and rapid pattern changes on flexible materials is also envisioned.
- **R2R Process Goals:** In the future, it is envisioned that R2R processes will include additive processes and conveyance technologies for brittle substrates such as glass and ceramics. Also, multi-material deposition processes will be matured for different devices and provide better adhesion, higher yield and improved quality. It is also envisioned that there will be more process applications for wide web (> 1 meter) flexible glass substrates. Finally, one goal is to have the capability of fast iteration from R&D to piloting/membrane upscaling to include modular facilities, material characterization, in-line sensing, and real-time process control.
- **Cost:** Many cost goals were envisions by the breakout group. This included new R2R processes that deliver low cost, very thin R2R substrates (about 1 millimeter thick) with sufficient

mechanical integrity for ease of handling (without wrinkles or breaks) and moderate temperature processing (150-300 ºC). For desalination applications, the cost is envisioned to be $0.25/cubic meter. For battery applications, the cost target for proton conducting membranes is less than $50/square meter.

Challenges and Barriers

On the discussion of challenges and barriers, key recurring themes are highlighted below. The full results of the discussion are provided in Table C-2 of Appendix E.

- **Materials and Substrates**: Material incompatibilities between additives, polymers, and curing processes prevent multiple process steps from being performed in-line. This includes difficulties in handling, transporting, and packaging brittle substrate and membrane materials. A lack of knowledge for R2R coating of porous substrates (infiltrating, non-infiltrating, and conformal) and degradation of R2R coating processes from material impurities reduce overall process performance. Low-cost functional nanoparticles of desired size distribution and methods to incorporate production of 2-D and cellulosic nanomaterials in R2R processes need further development.
- **Membrane Fabrication**: There is a poor understanding of functional material chemistry to maximize covalent bonding onto membrane surfaces. Methods to fabricate atomically thin membranes (e.g., graphene, boron nitride, etc.) are needed. Currently, membranes have insufficient selectivity and permeability and better deposition techniques for dense ceramics on thin polymers are needed.
- **Metrology and Process Control**: In-line metrology tools are needed to characterize membrane performance during manufacturing, provide better process control with adequate sensors, and provide real-time feedback during deposition processes. There is also no capability for in situ defect detection at line speeds or to characterize nanostructured materials over a large area. Improved measurement tools, computational tools, and fundamental models need to be developed. Better capabilities are needed for multilayer/multi-phase structural control and particle morphology control for optimal process performance.
- **Scalability**: Lack of equipment, process capabilities, and system integration coupled with process and design limitations that come with the use of shared institutional equipment prevent the scale-up of new technologies for R2R processing. Insufficient stability and durability for R2R equipment result in poor reproducibility of products in R2R processing. There is an inability to combine materials/multilayers for cost-effective mass production. The high costs for monomers/high temperature polymers and low market volumes result in fewer capital investments and resources for R2R manufacturing using these materials.

R&D Needs

On the discussion of R&D needs, key recurring themes are highlighted below. The full results of the discussion are provided in Table C-3 of Appendix E.

- **Materials and Substrates:** High-speed, low-cost R2R processes for functionalization of membranes for batteries and filter applications and functionalization methods for polymer substrates will benefit industry. R2R methods need to be developed for different membrane and substrate properties (e.g., flexible substrates, different particles sizes/shapes/distributions, and micro-structural control). Additive R2R processes (e.g., thin films, coatings, 3-D structures) for novel substrates (e.g., ceramics, glass, advanced polymers) and printable materials with designable properties (e.g., optical, mechanical, electrical, thermal, chemical) would be improvements to R2R processing. Further work is needed on binders, multi-materials fusion, surface activation, advanced mixing techniques, complex materials deposition, and techniques for metallizing a top surface and graded surface loading on porous substrates.

- **Membrane Fabrication:** Additional research will improve our fundamental understanding of membrane surfaces and topologies (e.g., surface energy, adhesion, fouling) and interactions between materials under various processing conditions. Development of novel polymer composite membranes, fast ion/electron-mixed conducting membranes, functional membranes without solvents, and defect-free thin film membranes will solve many of the problems associated with today's clean energy technologies. Methods need to be developed for increasing membrane selectivity (including modeling), producing stable, tunable membranes with hierarchical structures, and improving membrane flux, fouling resistance and robustness. R2R processes for solution deposition of ultra-thin (<10 nanometers) dense films and to fabricate discrete multilayer composites of different materials and dimensions are desirable.
- **Metrology and Process Control:** R&D is needed for high-throughput, rapid (<1 second) in-line characterization of the structure and morphology of buried interfaces and large-area characterization of molecular packing, phase segregation, pore distribution size, and conductivity of membranes including in situ / in operando monitoring and control. Additional development will provide an effective, rapid process for measuring membrane performance indirectly during manufacturing processes. Advanced multilayer simulation tools, databases and predictive analysis methods that link membrane structure, properties, and performance require improvement. Control of substrate alignment, registration, and surface roughness to a few nanometers (which is needed to achieve high-quality, thin, and uniform printed films) will enhance R2R processing.
- **Scalability:** Models to better understand how to use nanoscale materials processing to meet performance goals and materials growth pathways to achieve them will facilitate the adaptation of new materials to existing infrastructure. A capability is desired for rapid iteration of R&D to pilot scale with a flexible, modular infrastructure and a programmatic goal of 50% reduction in the cycle time from laboratory to prototype. A flexible, integrated membrane fabrication system along with adequate testing infrastructure will allow for material modifications to transition from conventional to high speed R2R processing more effectively. Process economics need to be properly evaluated and validated.

Priority R&D Topics

A set of priority topics for R&D emerged from discussions on the challenges and R&D needs identified for *Membranes and Substrates and Associated Functional Materials and Cross-cutting Applications*. These topics represent areas where a concerted effort in R&D could help overcome major material and manufacturing technology challenges. For each priority R&D topic, the breakout session participants focused their discussion on:

- Key challenges,
- Desired outcomes,
- Appropriate near-term and the long-term R&D steps,
- Performance goals and targets,
- Potential participants and roles, and
- Impact on clean energy industry.

The topics are summarized below and described in more detail in Figures C-1 through C-4.

- **Materials and Substrates:** Targeted R&D in materials and substrates will address the challenges in incorporating new functional materials into R2R processes. Also, the challenges involved in depositing high-temperature materials on low-temperature substrates will be investigated. This will yield new process tools that will benefit companies, both large and small, and identify rules and tools for membrane and materials selection.

- **Membrane Design and Fabrication for R2R Processing:** This effort will focus on identifying cost-efficient approaches and processes that facilitate transitioning from lab-scale to high-volume production without a loss in production quality**.** *In situ* high-quality process modeling tools for in-line QC and consistency will also be an area of interest. As a result, high-volume manufacturing with high-yield would be possible. In addition, new materials can be engineered for increased selectivity, durability, and reliability.
- **Metrology and Process Control:** This effort on metrology and process control focuses on the following capability gaps: the ability to predict properties/functionality in line; real time process control using existing technology; and technology that enables characterization over large areas. With new metrology and process control technologies and processes, it would then be possible to enable real-time measurement with a feedback loop to adjust parameters with minimum trial and error. This effort will also generate new models that can be used to determine the appropriate set of parameter combinations.
- **Scalability, Technology Transfer, and Scale-up:** This targeted R&D effort is designed to address technology scale-up challenges. There is a shortage of infrastructure access, trained personnel, and knowledge that takes a new process or technology from the laboratory environment to commercial scale. Industry engagement in this area is also challenging. Once these challenges are mitigated, products such as high-performance membranes can be produced at-scale. Not only will this establish a pathway for rapid transfer from R&D to pre-pilot to manufacturing scale, additional workforce training will also be possible.

Process and Equipment Needs for Transitioning Plate-to-Plate to R2R

This breakout group focused on three key topic areas related to the use of *Process and Equipment Needs for Transitioning Plate-to-Plate to R2R*:

- Topic 1: Vision and Goals
- Topic 2: Challenges/Barriers
- Topic 3: Priority R&D Topics

For each of area, one focus question and several additional questions were posed. Individual participant's views and responses were captured using a compression planning and brainstorming process. Highlights of discussions are outlined below; the full results of discussions are provided in the Appendix E.

Several themes emerged throughout the session, all of which are interconnected and contribute toward the vision of a cost-efficient approach to transition plate-to-plate processes to R2R, so as to achieve increased yield while benefiting from cost reduction, with targets such as increased production capacity by a factor of 10 times while reducing the cost to manufacture (including energy and labor) by 50%.

Vision and Goals

On the discussion of vision and goals, key recurring themes are highlighted below. The full results of the discussion are provided in Table D-1 of Appendix E.

- **Product Cost Reduction:** One vision is the production of cost effective sensors for building applications via R2R. Production capacity is envisioned to be increased by a factor of 10 times while the cost to manufacture (including energy and labor) is and reduced by 50%.
- **Process Cost Reduction.** The cost and time of R2R process testing and reiterating is envisioned to be brought to parity with plate-to-plate.
- **Quality Control/ Quality Assurance:** Significant improvement in quality and process control of the product is envisioned so that 10% to 20%material savings and reliability is achieved.
- **New Material and Process:** In the future, higher performance materials on R2R substrates (e.g., glass, material foils) with high temperature (>400ºC) deposition are required. New processes and materials are required to transition plate-to-plate process to R2R. Examples include new R2R processes that accommodate brittle materials for products such as ceramic solid-state batteries or new materials that enable plate-to-plate to R2R transition.
- **Cross-cutting and Cross-Pollination:** One goal is to transition technology and lessons learned from semiconductor processes to R2R. Also, a prototype facility is envisioned that houses both plate-to-plate and R2R equipment, control software, tools, etc., to enable the comparison of the two manufacturing processes and facilitate the transition to R2R.
- **New Processing Technology/Capability:** One goal is to achieve the ability to dial in process control parameters into a R2R process so that it will be equivalent to a plate-to-plate process. From the view of a high-technology silicon foundry, one envisions a R2R line that is operated in an affordable large chamber vacuum with traditional semiconductor fabrication processes such as atmospheric ceramic forming and continuous pulsed layer deposition.

Challenges and Barriers

On the discussion of challenges and barriers, key recurring themes are highlighted below. The full results of the discussion are provided in Table D-1 of Appendix E.

- **Slow Technology Transfer:** One key challenge is the time required to adopt new materials; it is currently too long. There is a need to decrease the time between discovery to production of new materials. It is also a challenge to transition a simple/small scale plate-to-place to R2R. This is rooted in a lack of understanding of scalability. The is a lack of a metric that indicates probability of success to scale-up a process.
- **Lack of New Technologies:** New technologies are required to facilitate and enable the transition plate-to-place processes to R2R. For example, current metrology and QC technologies are unable to provide insight to a product as it is being fabricated between rolls. New sensor technologies, measurement methodologies, and analytics to interpret QC and metrology data are needed. Also, current R2R technologies/equipment cannot produce products uniformly over large areas. Also, plate-to-plate forming techniques are not compatible to R2R.
- **Cost/Benefit Analysis:** The ROI in the transition from plate-to-plate to R2R is not well understood.
- **Lack of New Materials:** There is a lack of new materials to enable the transition from plate-to-plate to R2R. For example, in the transition of traditional stacking methods for batteries and fuel cells (and flow batteries) from plate-to-plate to R2R, new materials/chemistries will be needed to go from 10 ft^2/minute to 100 ft^2/minute. In addition, new roll materials "substrates" will be required to transition from plate-to-plate to R2R.
- **Lack of New Processes:** Process barriers are preventing easy transition from plate-to-plate to R2R. For example, the transition path for annealing techniques from plate-to-plate to R2R platform is unclear. There is also an insufficient understanding of the interfacial chemistry/physics of R2R materials under process and the process parameters in R2R process so that it can be dovetailed to a plate-to-plate process.

Priority R&D Topics

A set of priority topics for R&D emerged from discussions on the challenges and R&D needs identified for *Process and Equipment Needs for Transitioning Plate-to-Plate to R2R*. These topics represent areas where a concerted effort in R&D could help to overcome major process and equipment challenges. For each priority R&D topics, the breakout session participants focused their discussion on

- Key challenges,
- Desired outcomes,
- Appropriate near-term and the long-term R&D steps,
- Performance goals and targets,
- Potential participants and roles, and
- Impact on the clean energy industry.

The topics are summarized below and described in more detail in Figures D-1 through D-2.

- **Transition to Manufacturing:** This effort will address the lack of simple, small-scale process development and the long time it takes to adapt new materials and processes. This effort will establish infrastructure, new equipment and expertise to scale material from gram to kilogram scale and enable process modeling and escalation.

- **New Process Development:** This R&D topic addresses the lack of process parameters, physics, and chemistry understanding that is required to transition plate-to-plate processes to R2R. The result of this effort will be a faster transfer and scale-up from plate-to-plate to R2R.

Continuous Processing/Process Development Needs

This breakout group focused on four key topic areas related to *Continuous Processing/Process Development Needs*:

- Topic 1: Vision and Goals
- Topic 2: Challenges/Barriers
- Topic 3: Research and Development Needs
- Topic 4: Priority R&D Topics

For each area, one focus question and several additional questions were posed. Individual participant's views and responses were captured using a compression planning and brainstorming process. Highlights of discussions are outlined below; the full results of discussions are provided in the Appendix E.

Several themes emerged throughout the session, all of which are interconnected and contribute toward the vision of a cost-efficient R2R process that can fabricate products with multilayer coating at high speeds, with in-line measurement and process control that achieves in-line inspection, monitoring, and tracking at a 100% level. Additionally, identifying key metrics and pilot sites will be needed to facilitate mass adoption of new continuous process technologies.

Vision and Goals

On the discussion of vision and goals, key recurring themes are highlighted below. The full results of the discussion are provided in Table E-1 of Appendix E.

- **Multi-material Systems and Multilayers:** One goal is to develop an in-line multilayer (<1 micron) coating technology on thin films (5-10 microns) with yields greater than 95%. Also, a goal is to achieve high rate vacuum deposition processes (>100 angstroms/second) for complex or traditionally high temperature materials on low temperature substrates. Three dimensional structured coatings with both short and long range systems are also desired. A vision is also to achieve a cost reduction of 10 times to 50 times for membranes and multilayer substrates.
- **In-line Measurement and Process Control:** One vision is the capability to have alignment and print in a high-speed web at the nanometer to the micrometer scale. Also, in-line inspection, monitoring, and tracking at a 100% level are also envisioned. Other goals include, pattern metals and dielectrics to 50 nanometer linewidths by R2R processing within the next five years, sensors and real-time measurement tools for R2R process control that are demonstrated and validated, and in-line monitoring for all layers in air from the nanometer to the macro structure level for organic photovoltaic applications.
- **Scale-up and Commercialization:** In order to facilitate scale-up, one goal is to develop appropriate operation metrics to allow comparison between processes, equipment, and plants. Also, there is a vision to demonstrate disruptive, low-cost, high-performance, and environmentally-friendly processes with successful case studies. Further, the participants envisioned successful demonstrations of large area R2R growth of inorganic semiconductors (e.g., silicon, germanium III-V) on inexpensive substrates with comparable properties to wafers. Also, membranes and materials would enable doubling line speeds with real time analysis and reduced operational expenses. Further, it is envisioned that, in the future, there will be a process for rapid iteration between materials R&D and R2R technology deployment. Furthermore, another goal is to establish an agile manufacturing demonstration facility for R2R continuous processing, product development, and real-time metrology that integrates pre-process, process, and post-process requirements. For specific clean energy technology application, one goal is to

achieve OLED lighting panels at $10/kilolumen and greater than two times cost reductions in with less than 10^{-6} g/m^2 deposition per 24 hours. Also, there is a goal to optimize incorporation of carbon nanotubes into desalination membranes prepared by a R2R process.

Challenges and Barriers

On the discussion of challenges and barriers, key recurring themes are highlighted below. The full results of the discussion are provided in Table E-2 of Appendix E.

- **Multi-material Systems and Multilayers**: Both process conditions for multi-material composites and material sets for multilayer processes are often incompatible for individual applications. Defect management strategies in multilayer multi-roll processing, including prevention and detection of defects, rework, and solvent and film compatibility between processes are inadequate.
- **In-Line Measurement and Process Control:** There is an absence of high-speed, high-resolution in-line and *in situ* monitoring and QC as well as on-line defect repair, especially for micron- and submicron-scale structures and flaws. Contributing factors are inadequate sensors and techniques for continuous data handling for pressure, temperature, particle loading, etc. Capabilities are needed for direct patterning of functional materials at the micron scale and below and low-cost diagnostic tools, such as spectroscopy and imaging, to characterize the R2R product over different length scales and feature sizes. Polymer webs for thermal and tension control and methods to monitor volatiles for dual Teflon belt processes need further development to improve R2R continuous processing.
- **Predictive Modeling and Computational Tools:** Key challenges for effective predictive modeling and computational tools are insufficient understanding of process parameters, methods, and tools to correlate process conditions to final device performance. Research is needed to provide an understanding of interfacial interactions between dissimilar layers during coating operations and studies to characterize nanoparticle dispersion and self-assembly during continuous processing. Methods are required for model validation, parameter correlation, simulation tools, vehicles, and metrics to support process design from laboratory to production.
- **Scale-up and Commercialization:** Industry would benefit from having centralized expertise in R2R technology development and a pilot facility to transition from the laboratory scale to the production scale. There needs to be greater leveraging of the relative strengths of academia, industry, and government without challenges related to intellectual property. Additionally, connections need to be improved between the R&D and manufacturing communities. Materials and precursors for rapid, robust processing are insufficient which leads to poor mechanical and structural stability of materials fabricated by R2R processes. A multi-use development system (i.e., with unique process/sensors for each user) and available process data and metadata that are reproducible and curated do not exist. Multidisciplinary R2R processes and tools as well as the use of the existing infrastructure are insufficient for transitioning from conventional manufacturing to R2R. R2R methods used today do not achieve the same level of quality as in batch mode. This fact, coupled with the high cost of current technology development, leads to overseas manufacturing being more viable.

R&D Needs

On the discussion of R&D needs, key recurring themes are highlighted below. The full results of the discussion are provided in Table E-3 of Appendix E.

- **Multi-material Systems and Multilayers:** New methods and processes need to be developed for rapid surface treatments and surface functionalization specifically for use in membrane manufacture using R2R processing. A method for achieving controlled microstructure versus random microstructure is needed for application to multilayer systems. Methods and tools are required for high speed registration (<1 micrometer accuracy) of sequentially patterned/deposited features in R2R processing, including the combination of dissimilar materials and processes.

- **In-line Measurement and Process Control:** Sensors and in-line monitoring of appropriate variables for QC and process control are major R&D needs. Development of rapid scanning electrochemical probes that are fast enough to map large areas and non-contact sensors for temperature, bias, and roughness would benefit R2R processing. Non-destructive defect identification technologies and measurement techniques need developed in the near term (<10 microns in two years) and long term (<1 micron in five years). Applications such as "internet of things" for real-time monitoring and reporting of sensor data should be investigated for use with R2R processes.
- **Predictive Modeling and Computational Tools:** Predictive models for surface tension, solubility (beyond Hansen solubility), and density functional theory informed molecular dynamics would aid in new materials development. Research is required for *in situ* characterization of materials and development of models for nanoscale films and microstructures that correlate layer structure, material properties, stresses, adhesion, performance, and process parameters. Flexible R&D-scale systems with characterization tools are needed to develop process models using performance characteristics and system correlations that can be validated and used for scale-up of R2R technologies and processes.
- **Scale-up and Commercialization:** Flexible R2R tools for vacuum additive processes and a modular pilot test bench to validate the tools need to be developed. Industry desires a network that connect R&D to manufacturing and categorizes the best-in-class R2R capabilities, mapped to impact, that can lead to down-selection for an institute's/facility's focus. Parallel methods for high-speed printing/imprinting (rather than serial writing) and direct write strategies to print 2-D hybrid and inorganic materials at sub-micron resolution are research needs that will assist in scale-up of some current technologies. A facility is needed for the demonstration of thin-film (<5 micrometer) size patterning at line speed and evaluation of multilayer membranes at both pilot scale and production scale. This facility could also develop cost models for forecasting cost from laboratory to production.

Priority R&D Topics

A set of priority topics for R&D emerged from discussions on the challenges and R&D needs identified for *Continuous Processing/Process Development Needs*. These topics represent areas where a concerted effort in R&D could help overcome major material and technology challenges. For each priority R&D topic, the breakout session participants focused their discussion on

- Key challenges,
- Desired outcomes,
- Appropriate near-term and the long-term R&D steps,
- Performance goals and targets,
- Potential participants and roles, and
- Impact on the clean energy industry.

The topics are summarized below and described in more detail in Figures E-1 through E-4.

- **Scale-up and Commercialization:** This addresses the challenges of the lack of cohesive network for R2R researchers, manufacturers, and other key stakeholders and the lack of resource availability for the R2R community. This effort will therefore establish a manufacturing demonstration facilities that will be anchors for R2R innovation in the U.S.
- **Predictive Modelling and Computational Tools:** This R&D priority area addresses the challenge of the lack of available models for wet film deposition, structure-performance

relationship, and in-line measurement and process control, just to name a few. This effort will be in user-friendly models.

- **Multilayer Deposition:** This effort will address the lack of techniques for rapid thermal processing and poor control over final surface topography. The conclusion of this effort will yield process capabilities ranging from micro-scale to nano-scale patterning and deposition techniques for atomically precise surfaces.
- **Process Monitoring and Control:** This R&D priority area addresses capability gaps in sensors adapted to a R2R environment for new materials, closed-loop feedback for process control, and understanding of material properties. This effort will yield rules and tools for transitioning new materials from TRL-4 to TRL-7 and standards, protocols and procedures for users.

Appendix A: Agenda

ADVANCED MANUFACTURING OFFICE
HIGH VALUE ROLL TO ROLL (HV R2R) WORKSHOP
DECEMBER 2-3, 2015

WESTIN Alexandria
400 Courthouse Square
Alexandria, VA 22314
(703) 253-8600

AGENDA

Day 1 (December 2)	
8:00 – 8:30 am	REGISTRATION FOR ATTENDEES
8:30 – 8:40 am	Welcome **Mark Johnson**, DOE Advanced Manufacturing Office
8:40 – 8:50 am	DOE Remarks **David Danielson**, Assistant Secretary, DOE Energy Efficiency and Renewable Energy
8:50 – 9:10 am	Advanced Manufacturing Office: Introduction and Interest in High Value Roll to Roll **Mark Johnson**, Director, DOE Advanced Manufacturing Office
9:10 – 10:30 am	Panel Discussion on HV R2R **Invited Participants (Non-Governmental Organization SMEs)**
10:30 – 10:40 am	Breakout Session Instructions
10:40 – 11:20 am	BREAK
11:20 am – 12:20 pm	Breakout Sessions: ➢ Advanced Deposition Processing and Printing for ALD, Thin, Mid and Thick-film Size-Scale ➢ Unique Metrology and Quality Systems for Specific R2R Applications ➢ Membranes & Substrates and Associated Functional Materials and Technologies for Crosscutting Applications
12:20 – 1:20 pm	LUNCH
1:20 – 4:00 pm	Breakout Sessions (Continued):
4:00 – 4:30 pm	BREAK
4:30 – 5:15 pm	Plenary Session: Breakout Session Summaries
5:15 pm	Adjourn

Day 2 (December 3)	
7:30 – 8:15 am	REGISTRATION FOR ATTENDEES
8:15 – 8:50 am	Facility Public-Private Partnerships in AMO **Mark Shuart**, DOE Advanced Manufacturing Office
8:50 – 9:00 am	Breakout Session Instructions
9:00 – 9:30 am	BREAK
9:30 am – 12:15 pm	Breakout Sessions: ➢ Process and Equipment Needs Transitioning Plate to Plate to Continuous R2R Additive Process Technologies ➢ Continuous Processing/Process Development Needs (both plate and web based, on-web and post-process requirements)
12:15 – 1:15 pm	LUNCH
1:15 – 2:30 pm	Breakout Sessions (Continued):
2:00 – 2:30 pm	BREAK
2:30 – 3:15 pm	Plenary Session: Breakout Session Summaries
3:15 – 3:30 pm	Closing Comments **Mark Johnson and Mark Shuart**, DOE Advanced Manufacturing Office
3:30 pm	Adjourn

Appendix B: Workshop Participants

Name	Organization
Alexander Agapov	W.L. Gore & Associates
Balu Balachandran	Argonne National Laboratory
Joseph Barton	FuelCell Energy, Inc.
Brian Berland	ITN Energy Systems, Inc.
Ahmed Busnaina	Northeastern University
Rolf Butters	US DOE
Miko Cakmak	University of Akron
Isaac Chan	US DOE – AMO
Derek Chi Lok Cheng	Ballard Power Systems
Gregory Cooper	Pixelligent Technologies LLC
Arthur Cotton	General Electric
Joe Cresko	US DOE – AMO
Zissis Dardas	United Technologies Research Center
Panos Datskos	Oak Ridge National Laboratory
Donald Davis	Kent Displays,Inc.
James Dayton	Proton Onsite
Thad Druffel	Conn Center for Renewable Energy Research
Pavel Dutta	University of Houston
Dan Fleming	Western Michigan University
Lorraine Francis	University of Minnesota
Matt Fronk	Kodak
Nancy Garland	US DOE – FCTO
Alison Gotkin	UTRC
David Gotthold	Pacific Northwest National Laboratory
John Hamer	OLEDWorks LLC
David Hardy	US DOE – AMO
Tequila Harris	Georgia Tech
Greg Haugen	3M
David Johnson	PARC
Pooran Joshi	Oak Ridge National Laboratory
Debra Kaiser	NIST
Greg Krumdick	Argonne National Laboratory
Stephen Lehrman	National Nanotechnology Coordination Office
Henry Lomasney	SANDIA SOLAR TECHNOLOGY
Goran Majkic	University of Houston
Mike McKittrick	US DOE – AMO
Mohan Misra	ITN Energy Systems, Inc.
David Moore	GE Global Research
Steve Nunez	US DOE – AMO
Christopher Oldham	VaporPulse Technologies
Paul Otis	US DOE – Energy Information Administration
Kendra Pierson	Headband for Today
Mark Poliks	Binghamton University
Ranjeet Rao	Palo Alto Research Center

Name	Organization
Lee Richter	NIST
Andreas Roelofs	Argonne National Laboratory
Drew Ronneberg	SMI, Inc
Craig Sheppard	AIMCAL
Stephen Sikirica	Advanced Manufacturing Office - U.S. DOE
Dennis Slafer	MicroContinuum, Inc.
Venkat Srinivasan	Berkeley Lab
Christopher M Stafford	Materials Science & Engineering Division, NIST
Mahendra Sunkara	University of Louisville
Mark Taylor	Corning Incorporated
Robert Tenent	NREL
Catherine Thibaud-Erkey	United Technologies Research Center
Phillip Thomas	A1 Solutions
Alireza Torabi	FuelCell Energy
Michael Ulsh	National Renewable Energy Laboratory
Mario Urdaneta	Advanced Manufacturing Office
Brian Valentine	US DOE – AMO
Maikel van Hest	National Renewable Energy Laboratory
David Ventola	B&W MEGTEC
Gary Voelker	Miltec UV International
Conghua "CH" Wang	TreadStone Technologies, Inc.
James Watkins	University of Massachusetts
David Wood	Oak Ridge National Laboratory
Hongping Yan	SLAC National Accelerator Laboratory
Angelo Yializis	Sigma Technologies Int'l

Appendix C: Acronym List

2-D	2-dimentional
3-D	3-dimensional
AFM	Atomic force microscopy
ALD	Atomic Layer Deposition
AMO	Advanced Manufacturing Office
ASTM	ASTM, formerly referred to as the American Society for Testing and Materials
BIPV	Building-integrated photovoltaics
CRADA	Cooperative Research and Development Agreement
DFT	Density functional theory
DME	Dimethyl ether
DOD	U.S. Department of Defense
DOE	U.S. Department of Energy
EERE	Office of Energy Efficiency and Renewable Energy
EOL	End of Life
FAA	Federal Aviation Administration
FDA	U.S. Food and Drug Administration
FOA	Funding Opportunity Announcement
IP	Intellectual property
kWh	Kilo Watt hours
LBL	Layer-by-layer
LBS	Pounds
MOCVD	Metal Organic Chemical Vapor Phase Deposition
Mph	Miles per hour
MRL	Manufacturing readiness level
MWH	Megawatt Hours
NDE	Non-destructive Evaluation
NEC	National Electrical Code
NNMI	National Network for Manufacturing Innovation
NSF	National Science Foundation
O&M	Operating and maintenance
OEM	Original Equipment Manufacturer
OLED	Organic light emitting diodes
ORNL	Oak Ridge National Laboratory
QA	Quality assurance
QC	Quality control
Quads	quadrillion British thermal units (quads)
PV	Photovoltaics
R2R	Roll-to-roll
R&D	Research and development
RD&D	Research, development and demonstration
RFI	Request for Information

REI	Reactive ion etching
ROI	Return on investment
SBIR	Small Business Innovation Research
SME	Subject matter expert
SPP	Strategic Partnership Project
SS	Stainless steel
TRL	Technology readiness level

Appendix D: Panelist Biographies

Dr. Fabio Albano, Director of Research & Development, Xalt Energy and Energy Power Systems

Dr. Albano directs R&D for XALT Energy with a team of 20 scientists and engineers supporting one of the largest lithium-ion battery manufacturing plants in North America. Dr. Albano is a founding member of Energy Power Systems (EPS) and a co-founder of Sakti3, a battery start-up manufacturing solid-state lithium-ion batteries that was recently merged with Dyson. He has invented materials recipes for cathode, anode and solid electrolytes and demonstrated manufacturing processes for first usable prototypes. He has published work on the design and optimization of power supplies for wireless integrated microsystems, implantable medical devices, materials science, circuit design, and widespread sensing applications. This work has led to 16 issued patents in both North America and Europe, along with awards in France and Italy. Dr. Albano is fluent in four languages.

Dr. Paul D. Fleming, Professor, Western Michigan University

Dr. Fleming has taught courses in digital printing and imaging, electronic publishing, digital color imaging, computer graphics, digital video, paper physics, lithographic printing, colloid and surface chemistry, and many body theory. During his career, Dr. Fleming has: conducted theoretical and computational research in industry and academia; utilized many disciplines, including chemistry, chemical engineering, computer science, materials science, mechanical and petroleum engineering and physics; and performed research in digital printing and imaging, printability, coatings for nonimpact printing, theoretical and computational interfacial phenomena, molecular composites, thermodynamics of phase behavior and critical phenomena, fluid mechanics, flow through porous media, statistical mechanics, molecular simulations, percolation theory, phase behavior of polymer blends, nonlinear optics, multiprocessor computing, networking of computers and numerical methods. He has more than 20 years' experience in academia, 10 years in polymer/rubber industry, and 12 years in petroleum industry and is the author of over 350 publications and presentations.

Dr. Venkat Srinivasan, Staff Scientist, Lawrence Berkeley National Laboratory

Dr. Srinivasan is a Staff Scientist at Lawrence Berkeley National Laboratory (LBNL) and Deputy Director of the Joint Center for Energy Storage Research (JCESR, the battery "Hub"). His research interest is in developing next-generation batteries for use in vehicle and grid applications. Dr. Srinivasan and his research group develop continuum-based models for battery materials and combine them with experimental characterization to help design new materials, electrodes, and devices. In addition to his research, Dr. Srinivasan is interested in moving technologies to market and has been exploring ways to develop an ecosystem, focused on batteries, which will accelerate technology development. In this role, Dr. Srinivasan conceived the idea of CalCharge, a one-of-a-kind public-private partnership in energy storage. Dr. Srinivasan has previously served as the technical manager of the Batteries for Advanced Transportation Technologies (BATT) Program, as the acting director of the BATT program, as department head of the Energy Storage and Distributed Resources (ESDR) department at LBNL, and the interim director of the ESDR Division at LBNL. Dr. Srinivasan joined the scientific staff at LBNL in 2003 after postdoctoral studies at the University of California, Berkeley and Pennsylvania State University. He received his Ph.D. from the University of South Carolina in 2000. He is also the author of a battery blog titled, “This Week in Batteries” found at http://thisweekinbatteries.blogspot.com/

Dr. Brian B. Berland, Chief Science Officer, ITN Energy Systems

Dr. Berland serves as the Chief Science Officer for ITN Energy Systems. In this role, he directs both technology and business development activities with a focus on moving technologies from the laboratory to commercialization. Over the last twenty years, he has led research activities in flexible electrochromic and low-e window films, flexible lithium solid state batteries, redox flow batteries, alternate energy generation and storage materials, environmental barrier coatings, as well as gas and liquid separation membranes. He has been active in all aspects of product development for these technologies including cost-modeling, development of strategic partnerships, and definition of product performance requirements/market identification to support successful commercial transition and scale-up. Dr. Berland joined ITN in 1998, after completing his post-doctoral studies at the University of Colorado. He holds a B.S. in Chemistry from Carleton College and a Ph.D. in Chemistry from the University of Colorado.

Dr. David Wood III, Senior Staff Scientist, Oak Ridge National Laboratory

Dr. Wood is a Senior Staff Scientist, Roll-to-Roll Manufacturing Team Lead, Fuel Cell Technologies Program Manager, and UT Bredesen Center Faculty Member at ORNL, researching novel electrode architectures, advanced processing methods, manufacturing science, and materials characterization for lithium-ion batteries and low-temperature fuel cells, and has been employed there since 2009. He is a well-known energy conversion and storage researcher with an industrial and academic career that began in 1995. From 1997 to 2002, he was employed by General Motors Corporation and SGL Carbon Group, excelling at applied R&D related to automotive and stationary PEFC technology. Later work (2003-2009) at Los Alamos National Laboratory (LANL) and Cabot Corporation focused on elucidation of key chemical degradation mechanisms, development of accelerated testing methods, and component development. Dr. Wood received his B.S. in Chemical Engineering from North Carolina State University in 1994, his M.S. in Chemical Engineering from the University of Kansas in 1998, and his Ph.D. in Electrochemical Engineering from the University of New Mexico in 2007. He was part of two LANL research teams that won the DOE Hydrogen Program R&D Award for outstanding achievement in 2005 and 2009. He was also part of the Cabot Corporation Direct Methanol Fuel Cell team, which won the Samuel W. Bodman Award for Excellence in 2008. Dr. Wood was also the 2011 winner of the ORNL Early Career Award for Engineering Accomplishment and led a team that won both a 2013 “R&D 100” award and 2014 Federal Laboratory Consortium (FLC) award with Porous Power Technologies. He has 12 issued patents and patent applications, authored 39 refereed journal articles and transactions papers, authored two book chapters, and given 59 technical presentations. His personal Web of Science impact factor is 90.4. Dr. Wood manages an average annual ORNL budget of $9.5M related to hydrogen infrastructure issues, polymer electrolyte fuel cells, and lithium-ion batteries.

David Ventola, Director of Business Development, Babcock & Wilcox MEGTEC

David Ventola is Director of Business Development, EP at Babcock & Wilcox MEGTEC. David holds a B.S. and M.S. in Chemical Engineering and a MBA. Prior to Babcock & Wilcox MEGTEC, David spent most of his career in the roll-to-roll coating & converting Industry. Just prior to joining MEGTEC David spent 5 years at A123 Systems focused on lithium-ion battery electrode manufacturing.

Appendix E. Detailed Breakout Results

A. Advanced Deposition Processing and Printing for ALD, Thin, Mid and Thick-film Size-Scale

Vision and Goals

FOCUS QUESTION 1: *In the next five years, what goals would we like to achieve in Advanced Deposition Processing and Printing? What are some of the specific targets we would like to reach?*

Table A-1 summarizes comments on the Vision and Goals the participants would like to achieve in advanced deposition processing and printing.

Table A-1. Vision and Goals
Manufacturing Throughput / Yield
• Increase speed of R2R coating by 5× for batteries/capacitors • Increase speed of R2R by 10× (many orders of magnitude) for printed electronics (atomistic size) • Print solar rigs using old paper mills (that were used for daily newspaper printing) • Manufacturing yield: for semiconductors, the goal is to increase uniformity and yield by one order of magnitude
Reduce Manufacturing Cost
• Dollars/m^2 goal: increase speed at lower/same cost – this should be a performance metric • Develop a metric tied to cost of product • Improved cost of performance ratios -> sensor platform, that are attractive to industry/CMOS hybrid electric • Develop low cost advanced deposition processes with very high quality substrates • Prove DOE's goal of $1/watt can be beat (try to reach $0.5/watt – objective for solar R2R)
New Manufacturing Technologies and Processes
• Processing outside of the dry room • Achieve interconnected multi-layers in printing • Start with cheap precursors to films in one R2R step, combine steps, molecules to tune composition • Be able to detect structure/chemical composition at high throughput • Integrate ALD R2R processes into battery lines – coating highly porous substrates • For future electronics, reach below micron applications (e.g., imprinting) at the100 nanometer range patterning of dielectrics – using multilayer, more sophisticated materials • Print features/device architectures at one micron (within 5-10 years) • Expand role for R2R in monitoring wearable technologies (film, textiles) • Achieve mixing technology for slurry-based deposition • "Not just a price metric for tech adoption" – go below a micron with PRP processes using R2R – alternative to vacuum deposition nano-scale thick • Multi-layer pattern coating on thin substrates with scratching out

Barriers and Challenges

FOCUS QUESTION 2: *What are the key challenges/barriers to development and use of advanced deposition processing and printing? What are the problems we are trying to solve?*

Table A-2 summarizes participant comments on barriers and challenges to development and use of advanced deposition processing and printing. Workshop participants were asked to vote on the barriers and challenges they perceived as most important. The number of votes received (indicating participants' highest priorities) is shown by asterisks and the vote count in parentheses.

Table A-2. Barriers and Challenges
Deposition Processes/Equipment
• Effectively model processes while considering introduction of alternative materials/process routes (8)******** • Room temperature and pressure printing at the nanoscale linewidth and thickness (6) ****** • Adapting R2R for "small" batch operations, making them modular (4) **** • Processes for dry manufacturing, deposition of materials on a R2R platform with composition, thickness, and cost control (4) **** • Lack of new materials and processes for low temperatures (<200º C) (which are needed for polymer substrates) (2) ** • Very high fluid (polymer) coating technology (like "spin coating") for submicron patterning via imprinting (2) ** • Lack of processes to deposit brittle and reactive materials in devices (1) * • For submicron patterning, need a process to produce uniform thickness residue later (1) * • Quality deposition in range of environments (e.g., humid, dry, hot, cold) • R2R of brittle materials: the process is an "art" and makes integration slow • Low temperature deposition of complex "single crystal" oxide materials and functional (e.g., for solar cells, energy harvester, sensors, etc.)
Materials/Substrates
• Material needs vs. manufacturing technology capability – for example, dry rooms, fast drying without cracking, calandaring without destruction (TRL vs. MRL) (3) *** • Ref:For chemical or water separations/water., need Chemistries and membranes that are supportive of greater chemical selectivity and rates (2) ** • Lack of understanding the effect of processing on the orientation/distribution of fillers in composite material (2) ** • Poor ability to achieve high quality functional nanomaterials by R2R platform processing in low-cost substrates (2) ** • Near single crystal quality films on R2R substrate (e.g., graphene, III-V materials) (2) ** • Substrate uniformity and cleanliness (1) * • Materials supply – slight changes in raw materials cause large changes in products (1) * • Lack of high quality materials by R2R; lack of understanding of cost reduction/benefit by R2R • 24/7 energy: make a structural composite that stores energy, must last for building lifetime
Scalability/Flexibility – New Manufacturing Processes
• Scalability of new manufacturing methods (8) ******** • Implementing nanofabrication methods into R2R (3) *** • Demonstration of R2R ALD at practical line speeds (3) *** • Scaling and wasting different substrates and equipment (e.g., powder, web, glass, textile) (3) *** • Integrating tool/process scalability and flexibility into decision making all along the R&D path (1) * • Identifying proper equipment (up front) and steps for scaling up a multistep process (1) *

Table A-2. Barriers and Challenges

Fundamental Chemistry/Properties

- Assessing long term/lifecycle properties of materials and systems when working at the R&D/laboratory level (4) ****
- Understanding process/limitations at the fundamental level and origin of stochastic variability (3) ***
- Determining origins of stochastic variability in R2R for improved yield and scalability

Defect Identification/In-line Control/Monitoring

- In-line QC, or how to control the parameters of interest in real-time (7) *******
- Registering/aligning multiple layers of coatings/patterns in a reproducible way, for a wide range of features (nanometers to millimeters); need metrology and tooling (3) ***
- Lack of nanometer contacts methods to detect defects of various sizes on web conveyed substrates; defects may be in patterned features, electrical, optical, or other physical properties (1) *
- Minimizing defects from particles (dust) and substrate irregularities (without a clean room) (1) *
- Active identification and removal of inhomogeneities in fluid systems suitable for continuous processes

Printing

- Printing of vertically interconnected multi-layered device below one micron scale (8) ********
- Layer compatibility in “ink” formulation, thin and thin, and ITO replacement (3) ***
- R2R printing of flexible electronics and energy harvesting technology on non-traditional substrates (2) **
- Develop better user friendly models to simulate the printing process (1) *
- 3D printing R2R for composite materials, for example polymer and carbon filler for high thermal conductivity (dry process)
- Real printable graphene drying and curing, online monitoring, and layer smoothing
- Controlled printing/deposition of carbon nanotubes across the size scale

Other

- Lack of collaboration in a highly competitive environment (10) **********
- There is a need for accessible demonstration facilities containing state-of-the-art and cutting edge tools and technologies to enable companies to assess new technology without the upfront risk (3) ***
- Consistent energy policy that would drive and incentivize industrial innovation and investments (3) ***
- Connected technologies identified and developed in parallel
- Communication between early stage researchers and manufacturers

Cost Factors

- High cost of investigating new materials and processes; high costs of and limited access to tools; limited access to expertise; high cost of developing tools that do not yet exist (2) **
- Capital intense manufacturing and demonstration
- Absence of objective information regarding each production or material “aspect” to the overall unit cost

Applications

- Lack of low energy processes (1) *
- High amount of energy required to produce structural panels (e.g., requires catalysts, highly reactive chemistry, non-contact heating)
- Working with suppliers who are also trying to develop their products

R&D Needs

FOCUS QUESTION 3: *Drawing on the technical challenges identified, what critical R&D is needed to overcome the major challenges, address scalability, and advance new technologies in the area of Advanced Deposition Processing and Printing?*

Table A-3 summarizes discussions on the critical technology R&D that will be required for advanced deposition processing and printing.

Table A-3. R&D Needs
Cross-Cutting R&D
• New methods for "dry" deposition of sub-micron to millimeter scale thick films with controlled porosity and uniformities • High speed electrical, optical, and physical property in-line measurement for R2R • Laser disk cut write of patterns/properties on R2R for tunable materials • Support and connection of nano-materials technology to R2R manufacturing platform • Cost effective disposition systems for large area, low-cost development of single crystal-like materials
Clean Energy Technologies
• Feasible structural battery technology • Innovative R2R chemistries and processes with lower energy consumption • Efficient processing/curing methods
Printing Vertically Integrated Multi-Layer Device Less than one Micron
• Thin and smooth ink formulation • Analysis of the interphase composition, integrity, and durability of a vertically integrated multi-layer device • Fundamental understanding of how processing parameters effect structure and performance of final layer
Low Temperature Processes/Materials
• Processes that do not require special environments (e.g., low RH) and are environmentally friendly • Fundamental studies of photonic annealing • ALD precursors that enable low temperature deposition (namely metals and complex oxides) • Non-contact heating technology (to compliment R2R production processing) • Understanding of the molecular dynamics and flow behavior of low temperature processes/materials
Scalability of New Manufacturing Methods
• Making the pathways for inter-connects (printing and imprinting) • New scalable methods for compositionally controlled synthesis • Scalable processes for micron scale R2R patterning of functional materials • Identification of the key parameters that need focus/attention (minimize scale-up time and cost) • Connect small scale with commercial volume users for early vetting and practical scale-up development • Conduct research scale vs. pilot scale system study • Characterization of limitations of ALD in R2R processing
Effectively Modeling of Processes
• Processing manufacturing informatics and statistical modeling materials (applying the genome approach to processing/R2R manufacturing)
In-line QC Parameters
• Small-scale pilot line(s) with in-line metrology for developing improved coating uniformity and/or sub-micron features • Develop in-line QC methods at small scale, then evaluate on commercial machines in-situ for feedback and refinement

Table A-3. R&D Needs
• Use liquid based suspension for low temperature printing • Define “defect” for each application and/or sector • Determine availability of cost-effective characterization equipment and methods • New methods for high speed (greater than 1000 meters per minute) sensing of films and fluids
Other
• Educated, skilled, and talented workforce

Figure A-1. Low Temperature Processing (< 200ºC) for R2R of Functional Materials on Polymer Substrates

KEY CHALLENGES:	DESIRED OUTCOMES:
• Most synthetic routes for oxides require high temperatures (about 400ºC -500ºC) • For ALD, water from the atmosphere impedes high speed R2R • Develop process protocols for transferring materials into devices and systems	• Integration of materials usually processed at high temperature onto low temperature R2R platforms • Low temperature vapor deposition processes capable of high speeds • Properties from these processes that are equivalent to high temperature routes

	Metal Oxides Integrated onto Polymers (low temperature)	High Speed Conformed Coating of Porous/Textured Substrates
R&D Approach		
<3 years	• Investigate nanoparticle routes for deposition and low temperature fusion (control of ligands) • Fundamental studies of photonic annealing • Research to develop new liquid precursor routes	• Evaluate current technology for infrastructure of high speed R2R in the context on nanoscale printing (LBL, ALD) • Explore directed assembly (electrophoretic) • Process survey of liquid and vapor deposition techniques
>3-5 years	• Create engineered substrates that enable control of temperature during annealing • Investigate over a range of thicknesses • Demonstrate material properties are acceptable	• Demonstrate materials properties are acceptable • Defining the pathway to large format R2R of ALD, LBL, and/or directed assemble
Performance Goals and Targets		
Metrics	• More diverse set of materials integrated into R2R processes (flexible substrates) • New devices and applications enabled by new materials	• Confirm submicron, uniform coatings over large formats/areas • New materials available for R2R
Potential Participants and Roles		
Stakeholders	None Noted	

Impacts

Saves energy – High: Low temperature
Reduces carbon, wastes, emissions, water – High
Accelerates innovation – High: New materials in R2R
Reduces costs – High: Energy-based

Improves product quality –Medium
Improves competitiveness – High
Increases raw material efficiency/yields – Medium: Additive process

Figure A-2. Multi-Scale Manufacturing Process (using R2R) for a Structural Energy Storing PV/Battery System for a BIPV

KEY CHALLENGES:

- Prove feasibility (disruption) of a structural building envelope that stores energy
- Show that the PV surface can match/deliver the required electric power
- Design and demonstrate economics, scalability, and lifetime
- Develop a system design (architecture) model

DESIRED OUTCOMES:

- Prototype design of a structural battery
- Produced prototype that is validated as functional and structural
- Determined cost of resulting energy storage and potential value metric versus separated battery/PV system

R&D Approach

<3 years

- Design and theoretical model of prototype energy storing PV
- Prepare a mockup and test – to prove
- Functional performance (testing)
- R2R process integration demonstration/development
- In-process quality assessment (including sensor technology)

>3-5 years

- Prove lifetime performance
- Explore military and FEMA applications
- Incorporate into residential (BIPV) service
- Achieve \$0.50/watt (\$0.03/kWh) economics

>5 years

- Establish a technology advancing network (within the U.S.) that will sustain "cutting-edge" technology status as this disruptive technology matures

Performance Goals and Targets

Metrics

- Show that energy storing PVs provide a significant contribution to DOE's cost objectives related to grid parity by constructing a full scale version and generating field test data
- Show that a "structural battery"/PV can be produced with R2R that will be at least 25% cheaper than a "conventional battery"/PV system

Potential Participants and Roles

Stakeholders

Industry/R2R Users – Home builders, BIPV compositors and licensors, R2R machinery manufacturers
Industry/R2R Technology or Material Developers – Materials manufacturers (battery media, polymeric coating composites)
National Laboratories – ORNL
Academia – University of Louisville

Impacts

Saves energy – High: Storing energy within the solar PV structure
Reduces carbon, wastes, emissions, water – High
Accelerates innovation – High
Reduces costs – High
Improves product quality – Medium
Improves competitiveness – High
Increases raw material efficiency / yields – Low
Other – High

Figure A-3. Scalability of New Manufacturing Processes

KEY CHALLENGES:	DESIRED OUTCOMES:
• Making the pathways for inter-connects (printing and imprinting) • Identifying the key parameters that need focus/attention (minimize scale-up time and cost) • Characterization of limitations of ALD in R2R processing	• New scalable methods for compositionally controlled synthesis • Scalable processes for micron scale R2R patterning of functional materials • Scalability – connect small scale with commercial volume users for early vetting and practical scale up development

R&D Approach

<3 years

- Understand fundamentals of each of the steps in the process flow diagram
- Survey experts in industry and academia
- Define and understand functional requirements
- Current R2R state-of-the-art materials and modeling (through consortia and/or database)
- Availability of different facilities at larger scales for various applications – e.g., vacuum deposition, electronic wet, flexible substrate
- Identify gaps and research to develop expertise in both manufacturing technology and equipment and material supply
- Collaboration and coordinated infrastructure

>3-5 years

- Conduct R&D on emerging materials and applications
- Refine and update first generation "processes" per application
- Provide system relevant data for future developments and methodology

>5 years

- Validate two new processes
- Adopt sustainability model and membership model NMMI

Performance Goals and Targets

Metrics

- Defined and understood functional requirements
- Multiple established large scale facilities for various applications – e.g., vacuum deposition, electronic wet, flexible substrate

Potential Participants and Roles

Stakeholders

Industry/R2R Users – Final machine design and process application specifications
Industry/R2R Technology or Material Developers – Demonstrate and supply pilot
National Laboratories – Provide prototype process and equipment
Academia – Define metrics for prototype

Figure A-4. Printing Dense and Vertically Integrated Devices

KEY CHALLENGES:	DESIRED OUTCOMES:
• Development of micron scale and below patterning and direct printing processes for R2R • Development of micron scale and below alignment and registration • Vias and interlayer connectivity • Processing at ambient conditions	• Integrated devices by R2R • Improvements in patterning and efficiency

	R&D Approach	
<3 years	• Patterning directly into functional materials • Eliminate photo resist-based patterning • Direct printing of nonorganic device layers • Minimum of substrate processes • Candidates include nanoimprint using novel materials, template, and transfer based processes, direct assembly techniques, and self-assembly techniques	• Approaches include optical, electrical, or magnetic alignment, layer and layer alignment for multiple processes, templates, nanoimprint, layer bonding, connectivity, adhesion, and orthogonal solvent approaches
>3-5 years	• Extend laboratory processes to R2R tools • Extend patterning processes to deep-sub-micron scale • Metrology for nanoscale features on R2R platforms	• High speed optical alignment
>5 years	• None noted	• None noted
	Performance Goals and Targets	
Metrics	• Sub-micron scale integrated devices	• Sub-micron scale integrated devices
	Potential Participants and Roles	
Stakeholders	None noted.	

Impacts

Saves energy – High: Storing energy relative to silicon
Reduces carbon, wastes, emissions, water – High: Saves water relative to silicon
Accelerates innovation – Medium: Enable intelligent devices

Reduces costs – High: Potential cost savings for high volume devices
Improves product quality – Medium
Improves competitiveness – High: Enables new markets
Increases raw material efficiency / yields – Medium

Figure A-5. In-line QC

KEY CHALLENGES:	DESIRED OUTCOMES:
• Attaining uniform thickness • Existence of point defects • Scalability and producibility of manufacturing processes	• Achieving 100% assessment/mapping • Ensuring feedback control • Having only periodic instead of random defects Develop new integrated material systems

◆ R&D Approach

<3 years	• Assess available solutions and techniques • Customize the available techniques for desired in-line QC
>3-5 years	• Develop new methods for high/medium speed sensing
>5 years	• None listed

◆ Performance Goals and Targets

Metrics	• Short term: up to 100 feet/minute (speed) • Long-term: 2% resolution in height measurement

◆ Potential Participants and Roles

Stakeholders	Industry/R2R Users – Testing and evaluation of the concepts Industry/R2R Technology or Material Developers – Commercialize the measurement instruments National Laboratory & Academia – Create new enabling technologies for measurements Associations – Develop standards and specifications

Impacts

Saves energy – Medium: Less materials consumption
Reduces carbon, wastes, emissions, water – High: Less waste
Accelerates innovation – Low
Reduces costs – High: Increases yield and decreases waste

Improves product quality – High: Fewer failures
Improves competitiveness – High: Higher quality/less cost
Increases raw material efficiency / yields – Medium

B. Unique Metrology and Quality Systems for Specific R2R Applications

Vision and Goals

FOCUS QUESTION 1: *In the next five years, what goals would we like to achieve in metrology and quality systems for R2R applications?*

Table B-1 summarizes comments on the vision and goals the participants would like to achieve in metrology and quality systems for R2R applications.

Table B-1. Vision and Goals
Cross cutting Applications
• Adapt existing QC technologies to new material processing, with a focus on clean energy applications
Metrics
• Measure the financial impact of R2R on hydrogen fuel cell manufacturing via metrics such as ROI • QC metrics that incorporate the benefits of big data analytics
Measurement Needs and Technologies
• Increase the measurement frequency of rheology without significant cost increase • Achieve 10nm in-line profilometry in 100,000mm^2/min production
QC/QA
• Ability to produce current clean energy technology via R2R at higher quality • New techniques and technologies, at lower cost, that enable the detection of contamination in the z-axis • New QC techniques that enable faster areal quality measurements and contamination detection • Enable application of R2R QC in a harsh manufacturing environment (e.g., high temperature, high pressure) • High-accuracy detection, with a high-resolution (micron-scale) measurement capable sensor, of application-specific coating defects • Physics-based modeling to enable accelerated testing, which will integrate feedback and data measured *in situ* on the R2R line
Manufacturing Process and Control
• New control mechanism that includes corrective action feedback loops for fixing defects on the R2R line. These control mechanisms will need to be tailored to specific applications • Control systems with algorithms that optimize manufacturing throughput, both at the modular and the plant level • Automatic process control based on non-contact, in-line sensors. Control point depends upon parameter of interest, range of direct, and inferential sensors/sensing technology • In process contamination removal • In process automated inspect and repair • Improved understanding of the relationship between defect type/size, deposition layer, and/or device performance/lifetime. This will help answer questions such as “What is the defect?” and “What, in the process, caused the defect?”
Analysis and Simulation
• Improved predictive analytics, where collected data are used efficiently and effectively. Also, data formats are standardized • Increased use of the collected data by operators at the plant • New automation and plant control software that takes a holistic view of the manufacturing plant. This software will not only be integrated to each process, the users are able to use this software to simulate different manufacturing operations and modalities • Advanced forensics to identify specific cause(s) of the product defect

Table B-1. Vision and Goals
• Decrease start stop time, especially metrology/sensing used to help quickly optimize a line during start-up • New algorithm that guide the user to optimally apply QC/QA • Integrate R2R process control with enterprise response planning

Challenges & Barriers

FOCUS QUESTION 2: *What are the key challenges/barriers to development and use of advanced metrology and quality systems for R2R applications? What are the problems we are trying to solve?*

Table B-2 summarizes comments on the key challenges/barriers to development and use of advanced metrology and quality systems for R2R applications. Workshop participants were asked to vote on the barriers and challenges they perceived as most important. The number of votes received (indicating participants' highest priorities) is shown by asterisks and the vote count in parentheses. In some cases, the category headings received votes as well.

Table B-2. Challenges and Barriers
Scale-Up
• Lack of ability to translate and apply laboratory measurements to in-line production (3) *** • Insufficient production volume to justify the capital cost required for an original equipment manufacturers (OEMs) to use automated QC/QA (3) *** • Insufficient understanding and familiarity of the manufacturing process to assess the impact of process inputs on the energy use, quality and throughput of the manufactured product (4) **** • High cost to implement new process control (some cost drives are software implementation, sensors, and detectors) (3) ***
Metrics and Standards (1)*
• Standard operating procedure (SOP) for metrology and quality systems for R2R does not exit • There are insufficient standards for metrology and quality systems for R2R
Measurement Technology (3)***
• Lack of non-visual imaging/areal measurements in a R2R process (4)**** • Lack of high resolution inspection capability of patterns measured on the R2R process • Many existing measurement technologies are too slow or not amenable to a R2R process environment (4)**** • Current metrology sensors cannot identify defects at process speeds (1)*
Data Analytics (3)***
• Current analytical approach cannot handle the data volume • High capital expense is required to leverage data analytics; ROI is not clear.
Material Science
• Insufficient understanding of device physics; a better theoretical and empirical understanding of material behavior is needed (5) ******
Material and Process Connection (8) ********
• Process-defect-performance relationships are not known • Lack of knowledge of what kind of defects a QC sensor should seek • Performance of devices produced on current R2R lines are not well correlated to identified defects. New measurement methods are needed, or have been established in unrelated industries. • Lack of predictive analytics that correlates material behavior to R2R process to product performance • Capability of today's metrology sensor and techniques are inadequate. For known defects, metrology techniques are not available. For unknown defects, additional insights are needed for if or when metrology technique would apply (2)**

R&D Needs

FOCUS QUESTION 3: *Drawing on the technical challenges identified, what critical R&D is needed to overcome the major challenges, address scalability, and advance new technologies in R2R in the area of Metrology and Quality Systems?*

Table B-3 summarizes comments on the R&D needed to overcome the major challenges, address scalability, and advance new technologies in R2R in the area of Metrology and Quality Systems. Workshop participants were asked to vote on the barriers and challenges they perceived as most important. The number of votes received (indicating participants' highest priorities) is shown by asterisks and the vote count in parentheses.

Table B-3. R&D Needs
Performance Testing (3) * **
• Ability to correlate long-term performance for device with different key defect types
Manufacturing Process (1) *
• Make QC/NDE measurement methods compatible with high line speeds* • New materials and processes that lend themselves to corrective QC actions
Modeling and Analysis (4) ******
• Development of an empirical knowledge base • Identify unknown "killer" defect • Process step/unit operations modeling that captures physics (i.e., film deposition process) and assists in development of new QC/NDE methods.
Leveraging Lessons Learned
• Study advances and lessons-learned in other countries and bring them back to US • Transfer QC technologies from other disciplines / industries to R2R *
System Level Study (5)*****
• Conduct system level study of the R2R manufacturing line to understand the relationship between process control, device material physics, and metrology • Instead of study of specific defect or process, study the interconnect between material science, process science and device physics
Sensor Technology (2)**
• Develop in-line functional measurements (as opposed to dimensional measurements), techniques, and sensors* • Improve x-y resolution of existing metrology sensors* • Identify ways to implement new sensors and sensing technology at reduced cost
Cost Cutting
• Reduce costs of existing (sophisticated) measurement techniques
Industrial Commons
• Develop a "cradle-to-grave" design center for new QC and metrology control techniques / technologies

Figure B-1. System Level Study

KEY CHALLENGES:	DESIRED OUTCOMES:
• Manufacturing process control • Identify defects • System integration of process science, materials science and metrology	• Device specified performance • Minimize/eliminate (device) defects to increase yield • Optimize the line.

	Continuous Process & Defect Detection	System Integration & Optimization
R&D Approach		
<3 years	• Apply analytics and physics/simulation for defects classification	• Standardize
>3-5 years	• New techniques for defect recognition (that are not available now)	• Big data analytics
>5 years	• *None stated*	• *None stated*
Performance Goals and Targets		
Metrics	• Increase yield (50% over 5 years) • Increase performance (by 2-times over 5 years)	• Decrease cost by 50% over 5 years
Potential Participants and Roles		
Stakeholders	*None stated*	

Impacts

Saves energy – High
Reduces carbon, wastes, emissions, water – High
Accelerates innovation – High
Reduces costs – High
Improves product quality – High
Improves competitiveness – High

Figure B-2. QC Techniques

KEY CHALLENGES:

- Delineating the QC needs for R2R materials
- Need for areal or linear point measurements
- Making detection techniques compatible with line speed

DESIRED OUTCOMES:

- Enable Six-Sigma quality for R2R clean energy materials

R&D Approach

<3 years

- Identify QC methods that are readily available
- Adapt available methods to get resolution and line speed
- Design new systems or modify existing systems to achieve targets

>3-5 years

- Transfer technology from laboratory to industry with partners to facilitate
- Quantify the benefits of QC techniques in terms of reduced scrap, increased accuracy, and lower cost
- Develop new sensors and techniques where existing systems cannot meet the near to midterm targets

Performance Goals and Targets

Metrics

Near Term

- Solution based: <10 meter; 100 ft/min
- Vacuum deposition: 100 nm; 2 ft/min

Mid Term

- Solution based: ~1 meter; 300 ft/min
- Vacuum deposition: 10 nm; 20 ft/min

Potential Participants and Roles

Stakeholders

Industry/Users: Define needs, materials sets, and process conditions
Industry/Material/ Technology Developers: Provide SOP equipment and materials
National Laboratory and Academia: Pilot and scale demonstrations of exiting or innovative techniques
Associations: Conduct modeling and data handing

Impacts

Saves energy – High: (e.g., sintering QC not devices)
Reduces carbon, wastes, emissions, water – Medium
Accelerates innovation – High
Reduces costs – High

Improves product quality – High
Improves competitiveness – High
Increases raw material efficiency / yields – High
Other (Profitability)– High

C. Membranes & Substrates and Associated Functional Materials and Technologies for Cross-cutting Applications

Vision and Goals

FOCUS QUESTION 1: *In the five years, what goals, targets and characteristics would we like to achieve for membranes and substrates and functional materials and technologies for cross-cutting applications for R2R?*

Table C-1 summarizes participant comments on vision and goals that they would like to achieve for membranes and substrates and associated functional materials and technologies for cross-cutting applications.

Table C-1. Vision and Goals

Membrane Design

- Multilayered, functional, and tailored structure in ceramic/metal/polymer membrane systems for gas/liquid separations with uniform and predictable properties
- Ceramic/hybrid membrane systems for water/organic separations with catalysis to eliminate biofouling
- High-barrier, multilayer polymer membranes with various additives for improved performance and durability (e.g., fuel cells, batteries, redox flow batteries, electrolyzers, etc.)
- Low fouling and low energy membranes that are thinner, tailored to surface texture, and stable at all temperatures
- Better selectivity of membranes in operating environments
- Membranes for harsh and challenging operating/storage environments
- Multi-component, defect-free, thin (micron scale), cost-effective (at a component level) polymer-based membranes for applications such as dehumidification, electrocalorics, etc.
- Nanostructured membranes for energy and cost efficient water purification and desalination
- Printable nanocomposite membranes with tunable properties
- Computational model development hand-in-hand with fundamental experimental studies when developing new membrane systems

Substrate Development

- High temperature, low-coefficient of thermal expansion substrates
- Functional substrates through nanoparticle/phase alignment by external field to achieve enhanced through-thickness properties such as electrical conduction, thermal conductivity, ionic conduction, and high dielectric properties
- Direct coating of smart materials on porous substrates with multilayers (in-line, controlled penetration) and rapid pattern changes on flexible materials
- Temperature insensitive and environmentally-stable substrates
- Low water vapor transmission rate substrates

R2R Process Goals

- Better in-line QC, namely R2R leak checking of membranes and assembled parts
- Additive processes and conveyance technologies for brittle substrates such as glass and ceramics

- Multi-material deposition processes for different devices that provide better adhesion, higher yield and improved quality
- More process applications for wide web (> 1 meter) flexible glass substrates
- Process in-line coated membranes with electrode active material for lithium-ion battery separator applications
- R2R process for solvent free battery electrode production and solid state battery production
- Better materials design and manufacturing techniques
- Flexible R2R products for various products/applications
- Faster iteration from R&D to piloting/membrane upscaling to include modular facilities, material characterization, in-line sensing and real-time process control

Costs

- Low cost, very thin R2R substrates (~1 mm thick) with sufficient mechanical integrity for ease of handling (without wrinkles or breaks) and moderate temperature processing (150-300° C)
- Cost goal of $0.25/cubic meter for desalination applications
- Low cost alternatives for proton conducting membranes (today $400/square meter; needs to be <$50/square meter)

Barriers and Challenges

FOCUS QUESTION 2: *What are the key challenges/barriers to development and use of advanced membranes and substrates and functional materials and technologies for cross-cutting applications for R2R? What are the problems we are trying to solve?*

Table C-2 summarizes participant comments on barriers and challenges for development and use of membranes and substrates and functional materials and technologies for cross-cutting applications. Workshop participants were asked to vote on the barriers and challenges they perceived as most important. The number of votes received (indicating participants' highest priorities) is shown by asterisks and the vote count in parentheses.

Table C-2. Barriers and Challenges
Materials and Substrates
• Materials incompatibilities that prevent multiple process steps from being performed in-line (8) ******** • Difficulties in handling, transporting, and packaging brittle substrate and membrane materials (8) ******** • Poor understanding/knowledge for R2R coating of porous substrates (infiltrating, non-infiltrating, and conformal) (5) ***** • Lack of availability of low-cost functional nanoparticles of desired size distribution (5) ***** • Lack of methods to incorporate production of 2-D and cellulosic nanomaterials in R2R processes (3) *** • Incompatibilities between additives, polymers, and curing processes (1) * • Degradation of R2R coating processes and performance from material impurities (1) * • Inability to perform high-temperature annealing on low-temperature webs
Membrane Fabrication
• Poor understanding of functional material chemistry to maximize covalent bonding onto membrane surfaces (8) ******** • Insufficient membrane selectivity and permeability (4) **** • Lack of deposition techniques for dense ceramics on thin polymers • Lack of methods to fabricate atomically thin membranes (e.g., graphene, boron nitride, etc.)
Metrology and Process Control
• Poor process control and inadequate sensing (9) ********* • Limited real-time feedback pre-, through-, and post-deposition (4) **** • Lack of in-line metrology tools to characterize membrane performance during manufacturing (5) ***** • Lack of *in situ* defect detection at line speeds (4) **** • Lack of measurement tools, computational tools, and fundamental models (2) ** • Inability to characterize nanostructured materials over large area (1) * • Poor particle morphology control for optimal performance (1) * • Lack of multilayer/multi-phase structural control
Scalability
• Lack of equipment, process capabilities, and system integration (5) ****** • Process/design limitations with the use of shared institutional equipment (3) *** • Low market volumes and resultant high costs (chicken and egg: volume and capital) (2) ** • Poor reproducibility in R2R processing • Insufficient stability and durability for R2R devices • Inability to combine materials/multilayers for cost-effective mass production • Low yields • Poor defect control • Lack of capital investment and resources • High costs for monomers and high temperature polymers

R&D Needs

FOCUS QUESTION 3: *Drawing on the technical challenges identified, what critical R&D is needed to overcome the major challenges, address scalability, and advance new technologies in R2R in the area of Membranes and Substrates?*

Table C-3 summarizes comments on the R&D needed to overcome the major challenges, address scalability, and advance new technologies in R2R for membranes and substrates and functional materials and technologies for cross-cutting applications. Workshop participants were asked to vote on the barriers and challenges they perceived as most important. The number of votes received (indicating participants' highest priorities) is shown by asterisks and the vote count in parentheses.

Table C-3. R&D Needs
Materials and Substrates
• Functionalization methods for polymer substrates (nanoparticles, processes) (11) *********** • Additive R2R processes (e.g., thin films, coatings, 3D structures) for novel substrates (e.g., ceramics, glass, advanced polymers) (6) ****** • High-speed, low-cost R2R processes for functionalization of membranes for batteries and filter applications (5) ***** • Printable materials with designable properties (e.g., optical, mechanical, electrical, thermal, chemical) (4) **** • R2R methods for different membrane and substrate mechanical properties (e.g., flexible substrates, different particles sizes/shapes/distributions, microstructural control) (3) *** • Multi-materials fusion (membrane/substrate) (2) ** • Methods for surface activation via R2R (1) * • Advanced mixing techniques to enable R2R deposition of complex materials (1) * • Techniques for metallizing a top surface (allowing a thin-film to act as an electrode) • Binders/solvents that allow in-line coating of lithium ion anodes and cathodes with separation • Methods to enable graded surface loading (such as catalysts with graded platinum loading or gas diffusion layers with graded polytetrafluoroethylene (PTFE) loading) on porous substrates
Membrane Fabrication
• Methods for increasing membrane selectivity, including modeling to drive the engineering of polymers for increased selectivity (5) ***** • Membranes for CO2 capture (3) *** • Fast ion/electron mixed conducting membranes (3) *** • High-throughput, rapid (<1 second) in-line characterization of structure/morphology of buried interfaces (3) *** • Methods for producing stable, tunable membranes with hierarchical structures (e.g., integrated catalysts, mechanical supports) (1) * • Techniques for producing functional membranes while avoiding use of solvents or water (especially for batteries and filter media) (1) * • Techniques for producing defect-free thin films (1) * • R2R processes to fabricate discrete multilayer composites of different materials and dimensions for use in fuel cells, flow batteries, electrolyzers, and other applications (1) * • Novel polymer composite membranes (graded properties, multi-functionality, additions such as carbon nanotubes and graphene) (1) * • Improved membrane flux, selectivity, fouling and robustness (1) * • Membranes for removing liquids / natural gas condensates (methane/ethane/propane/butane) • Solution deposition techniques for ultra-thin (<10 nm) dense films • Techniques for properly functionalizing and geometrically defining membrane topology • Incorporation of reaction catalysts with electrode for reactive separation (e.g., propane to propylene) • Improved fundamental membrane surface understanding (e.g., surface energy, adhesion, fouling)

Table C-3. R&D Needs
Metrology and Process Control
• Better process understanding (e.g., deposition, drying, sintering, etc.) that is more scientific and less Edisonian (7) ******* • Effective, rapid process for measuring membrane performance indirectly during the manufacturing process (i.e., can chemistry/microstructure measurements be correlated to membrane performance?) (7) ******* • Control of substrate surface roughness to a few nanometers (needed to achieve high-quality thin uniform printed films) (3) *** • Understanding of interactions between materials, function, and processing conditions, especially for particles (3) *** • *In situ* and large-area characterization of all parameters, including molecular packing, phase segregation, pore distribution size, and conductivity (3) *** • Substrate alignment and registration (1) * • Water feed databasing and predictive analysis (linking membrane structure, properties, and performance to water feed speciation) (1) * • Advanced multilayer simulation tools • *In situ* and *in operando* monitoring and control
Scalability
• Rapid iteration from R&D to pilot with a flexible, modular infrastructure (6) ****** • Models to better understand how to use nanoscale materials processing to meet performance goals and materials growth pathways to achieve them (2) ** • Programmatic goal: 50% reduction in the R&D cycle time (laboratory to prototype) (1) * • Material modification to transition from conventional to high speed R2R processing • Flexible, integrated membrane system fabrication and testing infrastructure • Process economics evaluated and validated • New materials adapted to existing infrastructure • Improved fundamental membrane surface understanding (e.g., surface energy, adhesion, fouling)

Figure C-1. Materials and Substrates

KEY CHALLENGES:	DESIRED OUTCOMES:
• Incorporating functional materials into R2R processes • Depositing high-temperature materials on low-temperature substrates	• Process tools for small companies • Rules and tools for membrane and materials selection

	R2R Deposition Processes	Functional Materials in R2R
	R&D Approach	
<3 years	• Develop sputtering processes for low-temperature substrates • Model drying processes	• Improve understanding of layer interfaces in R2R processes • Model R2R processes
>3-5 years	• Develop molecular models of solvent drying • Recycle and/or reuse solvents • Identify/develop environmentally friendly solvents	• Develop fault-tolerant systems vs. multilayer coatings • Permanently add functionality to existing materials
	Performance Goals and Targets	
Metrics	• *Not discussed*	• *Not discussed*
	Potential Participants and Roles	
Stakeholders	*Not discussed*	

Impacts

Not discussed

Figure C-2. Membrane Design & Fabrication for R2R Processing

KEY CHALLENGES:	DESIRED OUTCOMES:
• Preserving high quality while transitioning from lab-scale to high-volume production • Process design for cost-effective high-volume scale-up • *In situ* high-quality process modeling tools for in-line QC and consistency	• High-volume manufacturing with high yield • Engineering of materials for increased selectivity • Increased durability and reliability at membrane and system level

	Enhanced Yield / Cost Reductions	Membrane Systems for High Durability and Reliability
R&D Approach		
<3 years	• Transition from laboratory scale to pilot scale with cost projections at future high-volume production • Develop tools for in situ process control and monitoring to maintain high yield	• Develop accelerated lifetime testing protocols to validate end-product requirements at lab/pilot scale • Develop and validate high-fidelity process modeling tools at lab/pilot scale
>3-5 years	• Transition from pilot scale to high volume production, with cost projections • Implement process monitoring and control tools for high volume production	• Develop accelerated lifetime testing protocols to validate end-product requirements at high-volume production scale • Develop and validate high-fidelity process modeling tools at high-volume production scale
>5 years	• Perform life-cycle analyses of product and manufacturing processes	• Implement data analytics and customer feedback for continuous process and product improvement. • Leverage adjacent markets for further cost improvements
Performance Goals and Targets		
Metrics	• 10-50× cost reduction • >95% yields	• 10× increase in durability compared to current state-of-the-art processes • Selectivity up to 5× • Permeance up to 10×

Potential Participants and Roles

Stakeholders

Industry/Users: Not discussed
National Laboratory : Not discussed
Academia: Not discussed
Associations: Consortia would advance the state-of-the-art in this area; single entities cannot afford to undertake such large efforts individually.
Other: Not discussed

Impacts

Saves energy – High
Reduces carbon, wastes, emissions, water – High
Accelerates innovation – High
Reduces costs – High
Improves product quality – Medium
Improves competitiveness – High
Increases raw material efficiency / yields – Medium/High
Other: Reduces dependence on strategic materials - High

Figure C-3. Metrology and Process Control

KEY CHALLENGES:	DESIRED OUTCOMES:
• Prediction of properties/functionality in line • Real-time process control using existing technology • Characterization over large areas	• Real-time measurement with a feedback loop to adjust parameters • No more trial and error • Model determines which combination of parameters needs adjustment • Nanoscale monitoring across a large area (multiscale analysis)

	Determine Relevant Properties	Develop Measurement Tools
R&D Approach		
<3 years	• Data collection and data mining for the parameters that need adjustment (e.g. roughness) • Capture common needs across applications and sectors (e.g., membrane defects, thickness, wettability, conductivity)	• Apply knowledge (lessons learned) from semiconductor industry (batch processing), printing industry, and other industries to process control for R2R. • Identify gaps where there are no measurements (nothing can be ported from other industries)
>3-5 years	• Create knowledgebase to build predictive model • Model validation and experimentation • Link parameters from feedback loop to ultimate functionality/performance of device	• Develop tools that can address quality and integrity of interfaces • Develop tools for 3-D processing • Address shortcomings in data collection speed, or tradeoffs (resolution/speed, for example) • Miniature R2R tools
Performance Goals and Targets		
Metrics	• Extensive measurement capability in-line • Measurement platform that can be attached to a coater/R2R system • Real-time measurements (perhaps it is reasonable to hit 50% within 5 years?) • Measurements connected to the control system	•
Potential Participants and Roles		
Stakeholders	Industry/User: Guide needs; define requirements Industry/Material/ Technology Developers: Address gaps National Laboratory : Data collection; develop instruments; developing standards Academia: R&D and implementation (computational, analytical, experimental) Associations: Define common needs; develop standards; disseminating information; standardization	

Impacts

Saves energy – Low
Reduces carbon, wastes, emissions, water – Medium: yield increase
Accelerates innovation – High: Models will be helpful for innovation
Reduces costs – Medium: Yield increase
Improves product quality – High: Process control
Improves competitiveness – Medium: QC produces a competitive advantage
Increases raw material efficiency / yields – Low: Optimization of system

Figure C-4. Scalability, Technology Transfer, and Scale-Up

KEY CHALLENGES:	DESIRED OUTCOMES:
• Performance limitations, high costs of processes, and ill-defined processes at commercial scale • Difficulties in engaging industry • Lack of open access to infrastructure, trained personnel, and knowledge	• Low-cost, high-performance membranes, substrates, and materials at scale • Decentralized institute (such as an NNMI) for scale-up, testing, and training • Established pathway for rapid transfer from R&D to pre-pilot to manufacturing

Decentralized Institute Creation & Execution

	R&D Approach
<3 years	• Define and build the structure and operation of an institute • Research and define the relevant R2R / scaling technologies and map to locations/applications/personnel needed • Commission existing center for "quick wins" • Identify consortia needs, participants, and scope • Build databases • Define intellectual property/open source operational mechanisms • Define business plan to ensure sustainability, including establishing metrics
>3-5 years	• Build and commission new R2R products and technologies • Demonstrate value through predetermined metrics and successful case studies • Incorporate testing, real-time algorithms, and metrology
>5 years	• Continuously update market/technology needs • Industrial Internet
	Performance Goals and Targets
Metrics	• Disruptive methods and processes that are adaptable to R2R manufacturing
	Potential Participants and Roles
Stakeholders	Industry/Users: Collaborate on R&D; participate in institute; define path to market; develop commercial strategy; provide market understanding National Laboratory : Perform cutting-edge research; provide facilities Academia: Train next generation; perform cutting-edge research; develop novel materials Associations: Flextech

Impacts

Saves energy – Medium
Reduces carbon, wastes, emissions, water – Medium
Accelerates innovation – High
Reduces costs – Medium/High
Improves product quality – Medium
Improves competitiveness – High
Increases raw material efficiency / yields – Medium
Increases time to market – High

D. Process and Equipment Needs Transitioning Plate-to-Plate to R2R

Vision and Goals

FOCUS QUESTION 1: *In the next five years, what goals would we like to achieve for Transitioning Plate-to-Plate to R2R, specifically for processes and equipment?*

Table D-1 summarizes participant comments on vision and goals for materials, processes, and/or equipment used to Transitioning Plate-to-Plate to Continuous R2R Additive Process Technologies.

Table D-1. Vision and Goals
Product Cost Reduction
• Produce via R2R cost effective sensors for building applications (e.g., sensors that measure gases for indoor air quality) • Reduce cost by a factor of 10 • Increase production capacity by 10 times and reduce cost to manufacture by 50%, including energy and labor cost
Cost Reduction
• Bring the cost and time of R2R process testing and reiterating in parity with plate-to-plate.
QC/ QA
• Improve quality and process control of the product to achieve 10-20 percent material savings and reliability
New Material and Process
• Higher performance materials on R2R substrates (e.g., glass, material foils) with high temperature (>400ºC) deposition • Transition low temperature plate-to-plate manufacturing process to R2R process(es) • New R2R processes that accommodate brittle materials for products such as ceramic solids state batteries • New materials that enable plate-to-plate to R2R transition. These materials can withstand higher temperature manufacturing environment, support high yield, and are environmentally sustainable • Integration of ALD coating with R2R processing of porous films for battery thin conformal costings • Plate-to-plate to rotating wheel based R2R process for ceramics on polymer substrates • New materials that enable transition of plate-to-plate process to R2R process • Low cost metallization of polymers through R2R process
Cross Cutting and Crossing Pollination
• Transition of technology and lessons learned of semiconductor processes (e.g. lithography, reactive ion etching (RIE), and metal organic chemical vapor phase deposition (MOCVD)) to R2R. • Prototype facility that house both plate-to-plate and R2R equipment, control software, tools, etc., to enable the comparison of the two manufacturing processes and facilitate the transition to R2R.
New Processing Technology / Capability
• Ability to dial in process control parameters into a R2R process so that it will be equivalent to a plate-to-plate process. • Affordable large chamber vacuum that contain an R2R line • Atmospheric ceramic forming technologies that are adapted to R2R • Continuous pulsed layer deposition that are adapted to R2R • Strategies/methods that enable proper and sufficient sensors/measurement devices on a web line to determine/define quality/material performance of rolled goods

Table D-1. Vision and Goals
• R2R that accommodates the manufacturing conditions of silicon foundry process • Deployment of multi-layer R2R manufacturing process on the factory floor

Barriers and Challenges

FOCUS QUESTION 2: *What are the key process and equipment challenges/barriers to effectively transitioning plate-to-plate to continuous R2R? What are the problems we are trying to solve?*

Table D-2 summarizes participant comments on barriers and challenges for developing the key process and equipment to effectively transitioning plate-to-plate to continuous R2R. Workshop participants were asked to vote on the barriers and challenges they perceived as most important. The number of votes received (indicating participants' highest priorities) is shown by asterisks and the vote count in parentheses.

Table D-2. Barriers and Challenges
Technology Transfer
• Lack of investment in and attention to the transfer of new/existing R2R processes to new application (1)* • Time to adopt new materials is too long. There is a need to decrease the time between discovery to production of new materials (6)****** • Lack of simple/small scale process development that can be transitioned to R2R. This is rooted in a lack of understanding of scalability. For example, for a new process, what yield does it need to first achieve before industry believes that it is scalable? (11)*********** • Qualification time is too long, thereby increasing the expense for new materials development and use • Capital investment and resource required is too high
Technologies (QC/ etc.)
• Insufficient technologies that "sees" and provide insight to the product as it is being fabricated between rolls. New sensor technologies, measurement methodologies, and analytics to interpret the data are all needed • R2R technologies /equipment will need to produce products uniformly over large areas. New methods to monitor/control the process are needed (2)** • Lack of technology that facilitates the transition from plate-to-plate to R2R. For example, plate-to-plate forming techniques are not compatible to R2R • New equipment/processing designs are required to achieve desired line speeds. • Lack of approach and strategy to integrate new construction materials into new production • New equipment technologies/innovation to enable the R2R process requirements are needed (1)*
Cost/Benefit Analysis
• Lack of clarity on the most cost effective/impact going from plate-to-plate to R2R
New Materials
• Insufficient investigation on the likelihood of substitution of traditional stacking methods for batteries and fuel cells (and flow batteries) with all R2R process. Are there materials/performance limitations that prevent this switch? Are new materials/chemistries needed to go from 10's ft^2/min to 100's ft^2/min? (4)**** • New roll materials "substrates" so that the transition from plate-to-plate to R2R is possible does not currently exist (3)*** • Ability to obtain quality ALD coating on porous substrates at viable line speeds, conditions and cost*
New Processes
• Lack of integration of annealing techniques to transition from plate-to-plate to R2R platform • Insufficient thermal management for low temperature processes (1)* • Insufficient understanding of the interfacial chemistry/physics of R2R materials under process (6)****** • Lack of process science • Lack of understanding of process?—remove "art" • Annealing concepts are currently uncorrelated to thermo-physical models to define material and process space • New material and processes to enable R2R production are required (1)* • Lack of process parameters understand for roll to roll processes (7)*******

Table D-2. Barriers and Challenges
• Design based tools do not exist

Figure D-1. Transition to Manufacturing

KEY CHALLENGES:	DESIRED OUTCOMES:
• Lack of simple, small scale process development • Time to adapt new materials and processes that decrease time from discovery to production	• Access to equipment that is open for all to use • Infrastructure, new equipment, and expertise • Capability to scale material from gram to kilogram to enable process modeling and escalation

	Small Scale Process Development	Process Development and Materials Scale-up
R&D Approach		
<3 years	• Coordinated infrastructure and expertise • Identify gaps and establish new facilities as needed • Establish small scale sheet to sheet and roll to roll capability	• Transition small scale to large scale • Adopt technologies as available • Refine and validate with experimental data
>3-5 years	• Establish capability to scale materials from grams to kilograms • Cast modeling • Business intelligence and value proposition	• Work with models, train and transfer knowledge • Accelerate testing procedures • Refine and validate with experimental data
>5 years	• *None stated*	• *None stated*
Performance Goals and Targets		
Metrics	• Better capabilities to evaluate small scale process development • Demonstrate the ability to transition from plate-to-plate to plate-to-plate with performance as good as P2P	• Better capabilities to scale materials • Transfer of process knowledge to industry
Potential Participants and Roles		
Stakeholders	Industry/Users: Provide input on process specs Industry/Material/ Technology Developers: Use new capabilities National Laboratory: Pilot facilities Academia: Serve as R&D facilities Associations: Facilitate information exchange and define the challenge for industry Other: Consortium to collaborate efforts	

Impacts

Saves energy – High: ability to efficiently develop process
Reduces carbon, wastes, emissions, water – Medium: ability to design environmentally sound material process
Accelerates innovation – High: new capabilities will speed things up
Reduces costs – High: decrease time will lower the cost of development

Improves product quality – Medium: Small scale process development enables understanding of high performance low defects
Improves competitiveness – High: Reduce time to market
Increases raw material efficiency/yields – Medium: process optimization will increase yields

Figure D-2. New Process Development

KEY CHALLENGES:	DESIRED OUTCOMES:
• Lack of process parameters understanding for R2R • Understanding the physics and chemistry of R2R material interfaces under process	• Predictive design of R2R products • Faster transfer from plate-to-plate and scale-up to R2R • Develop new products enabled by the characteristics inherent in R2R processing

	Predictive Design of R2R	Process Development
R&D Approach		
<3 years	• Baseline current technology and product development cycle • Screen available tests • Initiate the qualification of tests, with focus on those unavailable to R2R	• Develop understanding of difference between static and dynamic processes • Develop new processes (and materials) appropriate for R2R • Identify opportunities unique to R2R
>3-5 years	• Develop of tools in the suite for modeling R2R processes and materials • Finalize standard tests	• Verification and validation of models • Continue process and material development • Demonstrate products and applications leveraging unique capabilities of R2R • Embed sensors for cradle-to-cradle; active sensors that monitor health and can survive environment
Performance Goals and Targets		
Metrics	• Suite of tools (physical testing, modeling, standards, metrics, and protocols) for assessing suitability and translation from plate-to-plate to R2R. • Cut current time to transfer from plate-to-plate to R2R in half	• Fundamental and applied understanding of plate-to-plate manufacturing processes
Potential Participants and Roles		
Stakeholders	Industry/User: Define applications, problems Industry/Material/ Technology Developers: Define products needs National Laboratory : Define problems, research, development, and demonstration Academia: Innovation, R&D	

Impacts

Saves energy – Medium
Reduces carbon, wastes, emissions, water – Medium
Accelerates innovation – High
Reduces costs – Medium
Improves product quality – High
Improves competitiveness – High
Other benefits (explain) – High

E. Continuous Processing/Process Development Needs

Table E-1 summarizes participant comments on vision and goals that breakout group participants would like to see achieved for continuous processing and process development needs.

Table E-1. Vision and Goals

Multi-Material Systems and Multilayers

- Develop in-line multilayer (<1 micron) coating technology on thin films (5-10 microns) with yields greater than 95%
- Develop high speed multilayer coating of dissimilar / composite materials
- High rate vacuum deposition processes (>100 angstroms/s) for complex or traditionally high temperature materials on low temperature substrates
- Standard functionalization and roughness for substrates
- 3-D structured coatings with both short and long range systems
- Achieve a cost reduction of 10-50times for membranes and multilayers
- Develop a generic, continuous, and room-temperature deposition process for a variety of materials
- Develop R2R processes with high yield and low defect films for multiple layers and nanometer to micrometer sizes

In-Line Measurement and Process Control

- Capability to have alignment and print in a high-speed web at the nanometer to micrometer scale
- In-line inspection, monitoring, and tracking at a 100% level
- New emerging technologies that allow for fabricating complex structure in-line versus flat sheet
- Interactions with enterprise resource processing systems for effective operations measurements
- Develop in-line device performance / permeability QC at operating speeds
- Direct patterning and alignment of functional materials at micro-scale on continuous platforms for high-speed, high-resolution printing
- Pattern metals and dielectrics to 50 nm linewidths by R2R processing within the next five years
- Develop, demonstrate, and validate sensors and real-time measurement tools for R2R process control
- In-line monitoring for all layers in air from the nanometer to the macro structure level for organic photovoltaic applications
- Ability to incorporate discrete elements (i.e., chips) into R2R processes with high resolution

Scale-Up and Commercialization

- Develop appropriate operation metrics to allow comparison between processes, equipment, plants, etc.
- Fast dry-dry deposition of materials (at standard temperature and pressure) in air environment
- Demonstration of disruptive, low-cost, high-performance, and environmentally-friendly processes with successful case studies
- Large area R2R growth of inorganic semiconductors (e.g., silicon, germanium III-V) on inexpensive substrates with comparable properties to that of wafers
- Decentralized R2R pre-piloting sites with unique capabilities to accelerate the path to commercialization that address material/substrate fabrication, characterization tools, in-line sensing, and feed processing
- Develop membrane or material performance and double the line speeds with real-time analysis and reduced operational expenses
- Continuous processing using dual Teflon belts to develop insight into non-contact heating methods such as microwave and induction and perforated belt technology for vapor release
- Develop a laboratory process (non-continuous) to permit pre-screening process evaluation and build a demonstration line
- Qualify process and economics at the pilot scale and establish supply chain requirements
- Apply standard additive processes to brittle substrates as effectively as polymers
- Establish an agile manufacturing demonstration facility for R2R continuous processing and product development and real-time metrology demonstration that integrates pre-process, process, and post-process requirements
- Develop a process for rapid iteration between materials R&D and R2R technology deployment
- Achieve OLED lighting panels at $10/kilolumen and >2× cost reductions in barriers (for OLEDs) with $<10^{-6}$ g/m2 deposition per 24 hours
- Optimize incorporation of carbon nanotubes into desalination membranes prepared by a R2R process

Barriers and Challenges

FOCUS QUESTION 2: *What are the key challenges/barriers to continuous processing and process development for R2R applications? What are the problems we are trying to solve?*

Table E-2 summarizes participant comments on barriers and challenges to continuous processing and process development for R2R applications. Workshop participants were asked to vote on the barriers and challenges they perceived as most important. The number of votes received (indicating participants' highest priorities) is shown by asterisks and the vote count in parentheses.

Table E-2. Barriers and Challenges

Multi-material Systems and Multilayers

- Incompatible process conditions for multi-material composites (6)******
- Lack of compatible material sets for multilayer processes (2)**
- Lack of defect management strategies in multilayer, multi-roll processing (e.g., prevention and detection of defects, rework, etc.) (1)*
- Lack of solvent and film compatibility between processes (1)*

In-line Measurement and Process Control

- Lack of high speed, high resolution in-line quality control (10)**********
- Inadequate sensors and techniques for continuous data handling (e.g., pressure, temperature, particle loading, etc.) (9)*********
- Lack of capabilities for direct patterning of functional materials at the micron scale and below
- Absence of on-line defect repair
- Inadequate diagnostic tools to characterize the R2R product over different length scales and feature sizes
- Insufficient integrated, low-cost characterization tools and methods, such as spectroscopy and imaging
- Lack of in-line metrology for micron- and submicron-scale structures and flaws
- Lack of in situ process monitoring and controls
- Need for polymer webs for thermal and tension control
- Volatiles in the process ribbon (for dual Teflon belt processes)

Predictive Modeling and Computational Tools

- Insufficient understanding, methods, and tools to correlate process conditions to final device performance (10)**********
- Insufficient understanding of interfacial interactions between dissimilar layers during coating (7)*******
- Lack of processing studies of nanoparticle dispersion and self-assembly (1)*
- Lack of validation models, methods, vehicles, and metrics to support process scale-up (laboratory to production)
- Lack of lumped parameter correlation / simulation tools

Table E-2. Barriers and Challenges

Scale-Up and Commercialization

- Lack of centralized expertise in R2R technology (9)*********
- Inadequate materials and precursors for rapid, robust processing (4)****
- Lack of centralized pilot facility to transition lab-scale to production scale (2)**
- R2R methods used today do not achieve the same level of quality as in batch mode (2)**
- Relative strengths of academia, industry and government are not leveraged (government and industry have investment dollars, and academics have time/equipment resources for research) (2)**
- Challenges related to intellectual property (2)**
- Poor connections between R&D and manufacturing (1)*
- Insufficient use of the existing infrastructure to transition from conventional manufacturing to R2R (1)*
- Lack of multi-use development system, i.e., with unique process/sensors for each user (1)*
- Insufficient multidisciplinary R2R processes and tools (1)*
- Poor mechanical and structural stability of materials fabricated by R2R processes (1)*
- Current cost of technology can lead to manufacture overseas to be viable
- Lack of available process data and metadata that is reproducible and curated

R&D Needs

FOCUS QUESTION 2: *Drawing on the technical challenges identified, what critical R&D is needed to overcome the major challenges, address scalability, and advance new technologies in continuous processing and process development for R2R applications?*

Table E-3 summarizes participant comments on drawing on the technical challenges identified, what critical R&D is needed to overcome the major challenges, address scalability, and advance new technologies in continuous R2R processing. Workshop participants were asked to vote on the barriers and challenges they perceived as most important. The number of votes received (indicating participants' highest priorities) is shown by asterisks and the vote count in parentheses.

Table E-3. R&D Needs
Multi-material Systems and Multilayers
• New methods for rapid surface treatments (5) ***** • New surface functionalization R2R process for use in membranes for batteries, filtration, house wraps, and heat management (5) ***** • Controlled microstructure (vs. random microstructure) in multilayer systems (4) **** • Rules and tools to guide the combination of dissimilar materials and processes (3) *** • Methods for high speed registration (<1 um accuracy) of sequentially patterned/deposited features (2)** • Level planar layers (1)*
In-line Measurement and Process Control
• Sensors and monitoring of appropriate variables for quality control and process control (5)****** • Rapid scanning electrochemical probe (e.g., conducting atomic force microscopy (AFM) for electrochemistry, fast enough to map large areas) (3)*** • Non-destructive defect identification technologies (<10 microns in 2 years, <1 micron in 5 years) (2)** • Fundamental studies of wetting, spreading, and solidification of ink for high resolution printing (2)** • Internet of Things (IoT) applications for real-time monitoring and reporting of sensor data (`)* • Non-contact sensors for temperature, bias, and roughness (1)* • Non-destructive measurement techniques • Metrology tools to track drying
Predictive Modeling and Computational Tools
• Predictive models for surface tension, solubility (beyond Hansen solubility), and density functional theory informed molecular dynamics (8)******** • Reference / test structure useful for R2R (can be used to correlate coating/device performance) (5)***** • Modeling of drying at different length scales (4)**** • In situ advanced characterization for better understanding to assist modeling under R2R conditions (4)**** • Predictive modeling of R2R growth processes to enable new materials development (3)*** • Microstructural models correlating layer structure, material properties, performance, and process parameters (2)** • Validated process models for scale-up (2)** • Flexible R&D-scale systems with characterization tools to develop process-performance characteristics correlations and models (1)* • Neuromorphic architecture for multi-modal input process controllers • Models for nanoscale film and coatings including adhesion, stresses, etc.

Table E-3. R&D Needs
Scale-Up and Commercialization
• Flexible R2R tool for vacuum additive processes (11)*********** • Modular pilot test bench (9)********* • Network that connects R&D to manufacturing (6)****** • Imprint and direct write strategies to print 2-D hybrid and inorganic materials at sub-micron resolution (5)***** • Parallel methods for high-speed printing/imprinting (rather than serial writing) (4)**** • Categorization of best-in-class R2R capabilities, mapped to impact (can lead to down-selection for an institute's/facility's focus) (2)** • "One-off" pre-production simulation station for dual-belt Teflon R2R (2)** • Sub-micron feature size patterning at line speed (1)* • Demonstration of a building-integrated, R2R-built solar PV system (1)* • Demonstration of thin-film (<5 um) and multilayer membranes at pilot scale and production scale • Cost models for forecasting cost from laboratory to production

Figure E-1. Scale-Up and Commercialization

KEY CHALLENGES:	DESIRED OUTCOMES:
• Lack of cohesive network for R2R researchers, manufacturers, and other key stakeholders • Lack of resource availability for R2R community	• Anchor R2R innovation in the U.S.

Demonstration Facility

	R&D Approach

<3 years	• Establish manufacturing demonstration facilities for:
	  ○ Vacuum-based R2R processes
	  ○ Wet R2R processes
	• Identify key players (e.g., users, expertise) for a decentralized facility

>3-5 years	• Decentralized manufacturing institutes like a Fraunhofer Institute.
	Performance Goals and Targets
Metrics	• Unified network for transitioning from laboratory to production scale • Databases for R2R knowledge • Center of expertise for R2R technology
	Potential Participants and Roles
Stakeholders	None discussed.

Impacts

Saves energy – Medium
Reduces carbon, wastes, emissions, water – Medium
Accelerates innovation – High
Reduces costs – High
Improves product quality – High
Improves competitiveness – High
Increases raw material efficiency / yields – Medium

Figure E-2. Predictive Modeling & Computational Tools

KEY CHALLENGES:	DESIRED OUTCOMES:

• Legacy practices / code limitations
• Lack of available models for
  • Deposition of wet films
  • Solidification (solution or monomer)
  • Structural hierarchy by post-solidification processes (e.g., thermal, microwave, strain)
  • Structure-performance relationship
	• User-friendly models
	• New methodology tools inspired by modeling needs

Demonstration Facility

R&D Approach

<3 years

- Down-select categories for modeling (e.g., solid, liquid, etc.)
- Develop multi-scale models of physical and chemical processes during solidification
- Implement integrated metrology / hierarchical modeling loops
- Examples include: skin formation with concentration gradient; particular coating with liquid drying; or kinetically trapped structures

>3-5 years

- Develop structure-function relation models
- Utilize models to predict appropriate material/process solutions for an application

Performance Goals and Targets

Metrics

- Quantitative tools that industry can use to optimize R2R lines
- Models to enable materials and process design based on performance targets (i.e., materials genome-type solution discovery)

Potential Participants and Roles

Stakeholders

Industry/Users: Define applications/challenges to be addressed
National Laboratory : Perform metrology and modeling; provide computational power
Academia: Perform metrology and modeling
Associations: Facilitate collaborations
Other: Collaborate internationally

Impacts

Saves energy – High
Reduces carbon, wastes, emissions, water – High
Accelerates innovation – High
Reduces costs – High

Improves product quality – High
Improves competitiveness – High
Increases raw material efficiency / yields – High

Figure E-3. Multilayer Deposition

KEY CHALLENGES:	DESIRED OUTCOMES:
• Availability of functional and carrier materials • Lack of techniques for rapid thermal processing • Poor control over final surface topography	• Compensation for mechanical/chemical effects • Process capabilities ranging from micro-scale to nano-scale patterning • Deposition techniques for atomically precise surfaces

	Material/Process Compatibility	Process Feasibility at Production Scale
R&D Approach		
<3 years	• Research and develop flexible, compatible functional layers • Draw from FlexTech materials • Compile data for body of knowledge	• Identify barriers to high speed, continuous processing
>3-5 years	• Discover and develop new materials/carriers • Build databases	• Invent new technologies/approaches to address barriers
Performance Goals and Targets		
Metrics	• Performance of components (e.g., voltage, power, etc.)	• Performance of components (e.g., voltage, power, etc.)
Potential Participants and Roles		
Stakeholders	**Industry/Users:** Demonstrate proof of concept **Industry/Material/ Technology Developers:** Provide materials **National Laboratory :** Develop databases **Academia:** Build understanding of materials and processes **Associations:** Provide support (capital and other)	

Impacts

Saves energy – Medium (in-line processing)
Reduces carbon, wastes, emissions, water – Medium
Accelerates innovation – High (stakeholder input)
Reduces costs – Medium

Improves product quality – High
Improves competitiveness – High
Increases raw material efficiency / yields – High

Figure E-4 Process Monitoring and Control

KEY CHALLENGES:	DESIRED OUTCOMES:
• Lack of sensors adapted to a R2R environment for new materials • Lack of closed-loop feedback for process control • Insufficient understanding of material properties (need to know what to control and monitor in the R2R process)	• Rules and tools for transition of new materials from TRL 4 to TRL 7 • Libraries of standards and specifications • Protocols and procedures for users

Model Development

	◆ R&D Approach
<3 years	• Identify key parameters to measure per material/process combination • Identify appropriate sensors for measurement needs • Develop new sensors where necessary • Develop new monitoring techniques • Collect and publish "library" information
>3-5 years	• Develop models and protocols for predictive process control • Demonstrate and validate monitoring techniques on pilot facilities • Develop feedback-based self-healing coatings and functional layers
>5 years	• Ensure that outputs are included in TRL 7-9 phase development of standards
	◆ Performance Goals and Targets
Metrics	• For solution processing: 10 µm resolution (in x-y axes) and 1 µm resolution (in z axes) • For vacuum processing: 10 nm resolution (in x-y axes) and 1 nm resolution (in z axes)
	◆ Potential Participants and Roles
Stakeholders	**Industry/Users:** Prioritize applications **Industry/R2R Technology Developers:** Provide sensors, controls, and equipment **National Laboratory :** Perform basic research on sensors and techniques **Academia:** Perform basic research on sensors and techniques **Associations:** Lead and coordinate efforts; disseminate information

Impacts

Saves energy – Medium
Reduces carbon, wastes, emissions, water – Medium
Accelerates innovation – Low
Reduces costs – High
Improves product quality – High
Improves competitiveness – Medium/High

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