Quadrennial Technology Review 2015

Chapter 6: Innovating Clean Energy Technologies in Advanced Manufacturing

Technology Assessments

Additive Manufacturing

Advanced Materials Manufacturing

Advanced Sensors, Controls, Platforms and Modeling for Manufacturing

Combined Heat and Power Systems

Composite Materials

Critical Materials

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U.S. DEPARTMENT OF
ENERGY

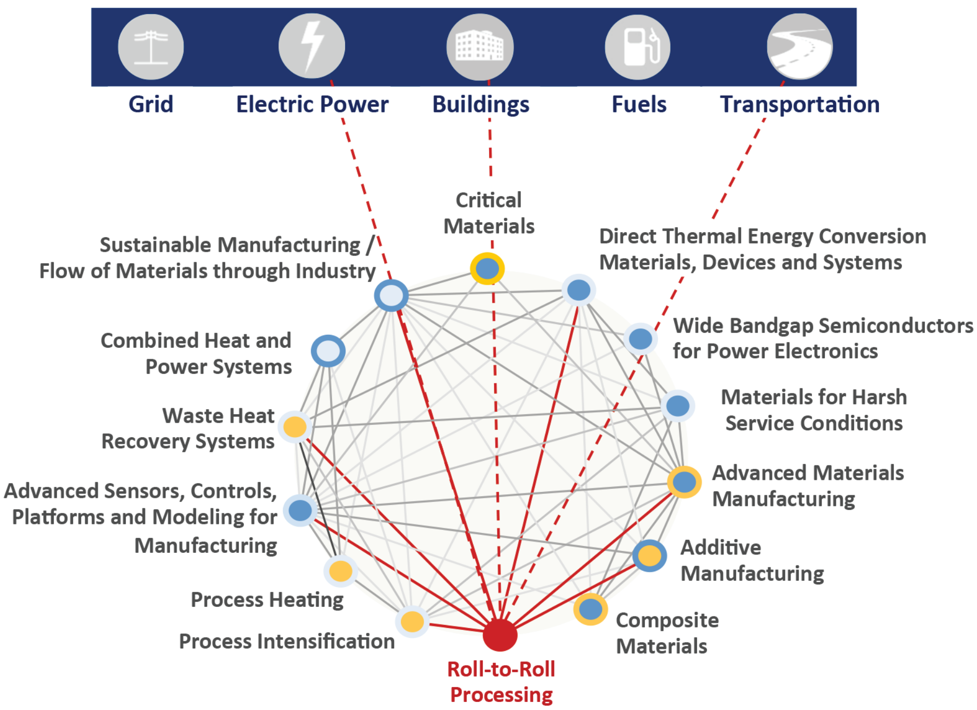
Quadrennial Technology Review 2015

Roll to Roll Processing

Chapter 6: Technology Assessments

NOTE: This technology assessment is available as an appendix to the 2015 Quadrennial Technology Review (QTR). ***Roll-to-Roll Processing*** *is one of fourteen manufacturing-focused technology assessments prepared in support of Chapter 6: Innovating Clean Energy Technologies in Advanced Manufacturing. For context within the 2015 QTR, key connections between this technology assessment, other QTR technology chapters, and other Chapter 6 technology assessments are illustrated below.*

Connections to other QTR Chapters and Technology Assessments



Representative Intra-Chapter Connections	Representative Extra-Chapter Connections
■ **Advanced Sensors, Controls, Platforms and Modeling for Manufacturing:** metrology and control systems for improved quality, defect detection, and throughput ■ **Process Intensification:** roll-to-roll for production of separation membranes ■ **Additive Manufacturing:** common technology needs for additive 2-D (roll-to-roll) and 3-D (additive manufacturing) printing technologies ■ **Direct Thermal Energy Conversion:** thermoelectric device fabrication via roll-to-roll ■ **Advanced Materials Manufacturing:** thin- and thick-film substrate production; multilayer alignment	■ **Electric Power:** flexible solar panels ■ **Buildings:** window insulation films ■ **Transportation:** battery electrodes and fuel cell membranes

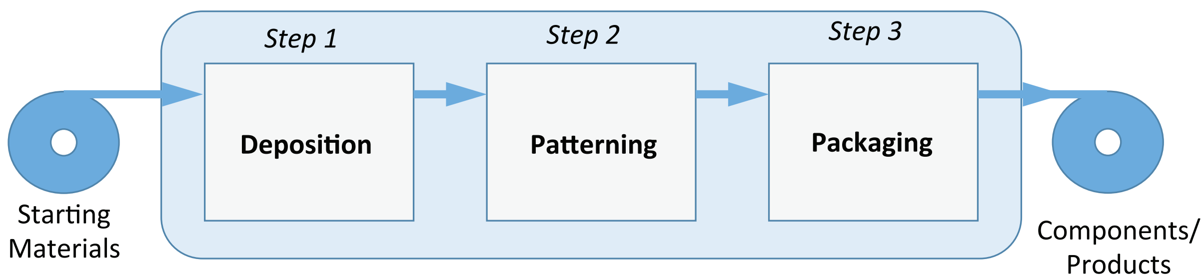
Introduction to the Technology/System

Overview of Roll-to-Roll Processing

Roll-to-roll (R2R) processing, also known as web processing or reel-to-reel processing, is an important class of substrate-based manufacturing processes in which additive and subtractive processes are used to build structures in a continuous manner. Related methods include sheet-to-sheet, sheets-on-shuttle, and roll–to-sheet; much of the technology potential described in this R2R technology assessment also applies to these associated substrate-based manufacturing methods. As an example of a R2R manufacturing operation, Figure 6.K.1 illustrates an idealized R2R processing concept for flexible electronics, where unit operations are sequentially performed on a "web" of flexible base material, which is ultimately wound onto a take-up roll.[2] Once the rolls of material have been processed, the take-up roll may be slit or cut into components or products, or used full-size in subsequent downstream operations. A wide range of intermediate and final products can be manufactured by R2R, where the number of process steps—and the specific unit operations of each process step—are dependent upon the requirements of the components/products.

Figure 6.K.1 Example R2R Processing Process Flow Schematic[2]

Credit: Adapted from A. Gregg, L. York, and Mark Strnad, "Roll-to-Roll Manufacturing of Flexible Displays," in G.P. Crawford (ed.), Flexible Flat Panel Displays, Wiley, 2005.



The series of sequential processing steps, along with the feed for input materials and winding finished materials, is called an R2R "line." Roller-based R2R lines are used when a continuous sheet or web can be conveyed on the line in an unsupported fashion. In addition to the web speed, the tension and position ("steering") of the web is typically controlled to ensure that the motion of the web across and around multiple rollers is done in a way that does not cause stretching or wrinkling of the web. Belt-fed lines are similar and are used when support of the web during processing is required (for example, during high-temperature process steps). Float lines allow long sheets of material such as glass to be processed while moving along a liquid surface. Finally, conveyors are used for cases where discrete parts are processed in a continuous fashion, such as for silicon photovoltaic (PV) wafers—although most processing of discrete parts is accomplished by using batch process methods. Table 6.K.1 provides an overview of the breadth of processes that are used in R2R processing lines.

Table 6.K.1 Examples of R2R Processes by General Type

Substrate Production	Continuous Coating
Casting, extruding, orienting, tentering, surface modification, cleaning, static mitigation, embossing, perforating, texturing	Roll coating, knife coating, die coating, spray coating, bath coating, sputtering, evaporation, vapor deposition, atomic layer deposition
Discontinuous Coating (Printing)	**Treatments and Post-processing**
Ink jet, aerosol jet, masking/etching, screen printing, flexographic printing, gravure	Drying, curing, sintering, annealing, chemical baths, cleaning, laminating, calendaring
Controls	**Conversion**
Speed, tension, steering, unwinding, winding, wrinkle mitigation, quality control, feedback/process control	Cutting, slitting, rewinding, packaging

A critical aspect of the use of R2R processing to create one or more functional layers is the choice of substrate. Substrates that are used in R2R processing may be made of a variety of materials, depending on the application and processing steps involved in fabrication. Mechanical, chemical, and thermal properties all may be important in the choice of substrate. Plastic films are desirable for their transparency, flexibility, and toughness but are often susceptible to degradation and dimensional distortion at high temperature. Where transparency is not required, stainless steel foils may be chosen as they tolerate higher temperatures than plastics. Other materials, such as aluminum and copper alloys, papers, flexible glass, fabrics, and nonwoven materials, are also commonly used substrates. Three common substrate materials are compared in Table 6.K.2.

Table 6.K.2 Comparison of R2R Substrate Materials[3, 4]

	Plastic Film	Stainless Steel Foil	Flexible Glass
R2R processing compatibility	Excellent	Excellent	Challenging
Mechanical stiffness (Young's modulus)	<5 GPa	200 GPa	70 GPa
Fracture toughness	1–5 MPa√m	62–280 MPa√m	0.5–0.8 MPa√m
Safe bending radius	4 cm	4 cm	40 cm
Visual transparency	Depends on polymer	None	Excellent
Coefficient of thermal expansion	72–198 x 10^{-6}	13–20 x 10^{-6}	1–10 x 10^{-6}
Thermal conductivity	0.1–0.3 W/m-°C	5–11 W/m-°C	0.7–1.5 W/m-°C
Maximum processing temperature	<300°C	1000°C	600°C

Given the breadth of products that do or can use R2R processing, a wide range of length scales—be it layer thickness, printed feature size, or surface roughness—must be able to be controlled and inspected. R2R equipment needs to support formats sufficient to meet nano-scale (e.g., atomic layer deposition (ALD)), small-scale (e.g., microelectronics thin and thick film), medium-scale (e.g., windows and window films), and large-scale (e.g., membranes for biofuel and natural gas processing) fabrication.

New manufacturing technologies are often seen as replacements for older processes that may be (for example) less energy efficient, lower yield, or lower throughput. R2R processing can be used as a drop-in process replacement in some cases; however, it is more commonly used as an enabling technology for new products. An R2R processing platform is often the only viable way to make a sheet or roll at high volume and at an acceptable cost. Thus, it is a critical manufacturing technology for many new clean energy and energy-efficient products.

Role of R2R in Addressing Environmental, Economic, or Security Challenges

R2R is a manufacturing method, not a material or product. Thus, its critical role is enabling the cost-effective production of high-value products that can address energy, environmental, and security challenges. In addition, further development of energy-efficient, low environmental impact, and lower cost R2R production processes can have significant economic and environmental impact within the manufacturing sector. Tools and equipment used in R2R processing often use less energy (per unit area of manufactured roll) for a much shorter duration relative to conventional manufacturing processes. For example, inkjet technology benefits include the reduced consumption of expensive materials, the reduction of equipment size and floor space compared with sputtering/photolithography, and low processing temperatures that permit the use of lower-cost plastic substrates.[5] For the manufacture of printed flexible organic photovoltaics (OPV) used in solar cells, the VTT Technical Research Centre (of Finland) has developed an R2R wet deposition printing process that enables environmentally benign production along with efficient material and energy use.[6]

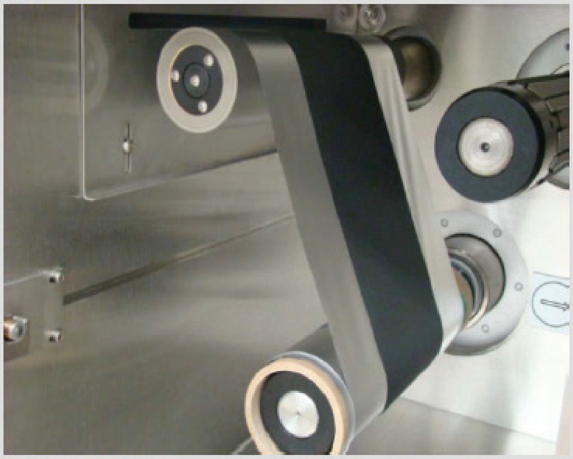
Some examples of mature markets for R2R products include print media, textiles, packaging, adhesive tape, magnetic tape, cling wrap, filter media, paper products, medical/hygiene products, and chemical and physical separations. The R2R concept is also applicable to some of the most advanced products in the world, many of which are of high interest to the U.S. Department of Energy (DOE) and other federal agencies. Examples of products with clean energy applications that have been produced by R2R processing techniques include the following:

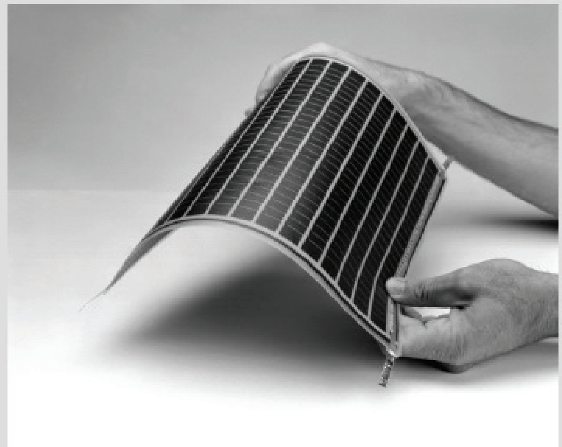
- **Flexible electronics:** Super-capacitors, electronic circuits, radio frequency identification (RFID) tags and labels (such as Smart Labels / Smart Tags that includes chip, antenna, and bonding wires), organic light-emitting diodes (OLEDs), displays, sensors
- **Flexible PVs:** Copper-indium gallium-selenide (CIGS), cadmium telluride, and other flexible PV products (Figure 6.K.2)
- **Printed/flexible thin-film batteries:** Laminar lithium-ion batteries (Figure 6.K.2)
- **Fuel cells and electrolyzers:** Planar solid oxide fuel cells, proton exchange membranes (PEMs), membrane electrode assemblies (MEAs), and gas diffusion media
- **Multilayer capacitors:** High-frequency dielectric capacitors for power conversion[7]
- **Thick-film sensor materials:** Temperature sensors, positioners, negative temperature coefficient thermistors, piezoelectric lead zirconate titanate actuators, active/passive transducers, selective gas sensors
- **Fabrics:** Clothing textiles and fiber reinforced mats
- **Anti-static, release, reflective and anti-reflective coatings:** Glass, Mylar®, polyethylene
- **Barrier coatings:** Thermal and environmental barrier layers
- **Building products:** Window films (electrochromics, reflectives, etc.), composite structural members
- **Chemical separation membranes:** Reverse osmosis membranes, catalyst membranes, gas separation membranes

Two examples of flexible thin film products made by R2R processing techniques are shown in Figure 6.K.2.

Figure 6.K.2 Examples of R2R Products: Battery Electrode (left) and Thin Film Photovoltaic Module (right)

Credit: (Left) David Wood, Oak Ridge National Laboratory; (Right) Warren Gretz, National Renewable Energy Laboratory





Technology Assessment and Potential

This R2R technology assessment reviews current state-of-the-art technologies, clean energy applications, and industry investments to advance R2R processing in the areas of metrology, equipment, carriers/webs, substrate materials, process improvement, alternative applications, and other possible innovations. These research efforts will serve to enable and maintain competitive R2R processing for domestic U.S. industries.

Recent Areas of Advancement

Both industry and the research community have pursued advancements in R2R processing, including new and improved processes as well as the application of those processes to new material sets. Several examples of recent advancements, demonstrating the breadth of technologies and markets that research and development (R&D) in R2R processing is helping to advance, are described below:

- **High-resolution printing:** A wide range of processes have been developed to meet the challenges of high resolution printing. Whereas in the 1970s one was scarcely able to print 25 µm wide lines and traces on a 250 µm thick substrate, investigators can now routinely print sub-200 nm features in a continuous web.[9]
- **Flexible glass substrates:** Corning, Inc. has developed flexible glass substrates with excellent surface quality, transparency, and high-temperature processing stability. Among other advantages, Corning has identified a potential 50% process cost reduction for these substrates through the use of R2R processing in manufacturing.[10]
- **Semiconductor thin films:** Researchers at the University of Louisville have developed a low-cost, low-energy intense pulsed-light treatment for rapid thermal annealing of materials, including cadmium sulfide (CdS) and cadmium telluride (CdTe).[11]

- **High-efficiency solar cells:** The current world record for CIGS PV cell efficiency is 21.7%, established in September 2014 by Stuttgart's Centre for Solar Energy and Hydrogen Research (ZSW).[12, 13] One technology that could make CIGS thin film production faster and more economical has been developed by engineers at Oregon State University in the United States and Yeungnam University in Korea. They are using continuous flow micro-reactors to replace current sputtering, evaporation, and electrodeposition techniques, which are slow and often require major vacuum systems.[14]
- **Single chip integration:** R2R has been used in single chip integration for smart labels (RFID tags and antennas), such as products offered by Muhlbauer High Tech International.[15]
- **Membranes for carbon dioxide (CO_2) capture:** Advances in CO_2 capture membranes fabricated with R2R processing have shown improvements in selectivity and permeability by improving bonding on novel active layers with polymer substrates.[16]
- **Thin-film transistors (TFTs):** Scientists at Hewlett-Packard Laboratories and Iowa Thin Film Technologies are developing large-area arrays of TFTs on polymer substrates using R2R techniques. Their approach combines plasma deposition and etching with self-aligned imprint lithography (SAIL).[17]
- **Organic photovoltaics (OPVs):** Several research groups have pursued advances in R2R processing for OPVs, including simultaneous multilayer coating,[18] aqueous processing,[19] and advanced drying and post-processing.[20] Two extensive reviews of R2R process demonstrations and research needs specific to OPVs are also available.[21, 22]
- **Membrane electrode assemblies (MEAs):** Cost reduction of fuel cell MEA materials is a key goal of DOE's Fuel Cells Technology Office (FCTO). Several recently completed cost-shared public-private R&D projects have achieved significant improvements in cost and quality of these materials as follows:
  - Ballard Material Products (now AvCarb, Inc.) reduced gas diffusion media costs by 55% through improved process control, increased production roll length and width, and empirical modeling to improve manufacturing efficiency.[23]
  - Gore Creative Technologies demonstrated sequential multilayer coating of highly dissimilar materials that would enable a 25% reduction in PEM MEA cost.[24]
  - BASF demonstrated a 30% decrease in platinum loading without loss in performance from the implementation of new coating techniques and a novel in-line scanning X-ray fluorescence (XRF) process for process control.[25]

Brief descriptions of R2R processes that encompass some of the aforementioned areas of advancement are provided in Table 6.K.3. The technologies listed are intended to be representative of current work, not exhaustive.

Table 6.K.3 Existing R2R Processes and Key Applications

R2R Process	Description	Example Applications
Vacuum deposition	Vacuum coating methods involve the deposition of materials onto a solid surface in a vacuum environment (i.e., at a pressure well below atmospheric pressure). Examples of vacuum deposition processes that can be successfully implemented in R2R processing include evaporation, sputtering, and chemical vapor deposition (CVD). Multilayer sputtering systems are common. During vacuum deposition, the entire substrate roll is loaded into the vacuum system, where different materials can be sputtered or evaporated onto the substrate without cross-contamination. This is more difficult in CVD, where reactive gas barriers are needed.[26]	Multilayer electrodes; supercomputer tape; thin film solar cells; OLEDs
Gravure	Gravure is a printing process that involves engraving the image onto a cylindrical image carrier.[27] The entire patterned cylinder is covered with ink, then the excess ink is removed with a doctor blade, leaving ink only within the recessed motif areas, as shown in the upper left of Figure 6.K.3. The plate cylinder is brought into contact with the impression cylinder to transfer the ink to the substrate via capillary action.[28]	Product packaging; print media
Flexographic printing	During flexographic printing, the substrate material is fed between the inked printing plate and an impression cylinder, transferring the image as the substrate passes through. Flexographic printing is essentially a modern version of a letterpress.[28]	Food packaging
Flatbed screen printing	In flatbed printing, a squeegee moves across a mesh design screen, forcing ink through the screen and onto the substrate, as illustrated in the bottom left of Figure 6.K.3. Generally, relatively thick wet layers can be achieved (10–500 μm).[29]	Displays; packaging
Rotary screen printing	In rotary screen printing, the substrate moves through rollers past a squeegee, forcing ink through a cylindrical design screen onto the substrate. This process is illustrated in the lower right of Figure 6.K.3.	Packaging; clothing
Imprint or soft lithography	In soft lithography, an elastomeric stamp, mask or mold is used to fabricate features on a substrate—typically at the micro- or nano-scale. Polydimethylsiloxane (PDMS), a silicone material, is most commonly used as the elastomeric mold material as a result of its chemical inertness, low cost, low toxicity, and biocompatibility. A related method is self-aligned imprint lithography (SAIL), in which multiple mask levels are imprinted as a single three-dimensional structure, avoiding the need for sample alignment.	Solid state lighting; displays; green building products; lab-on-a-chip; microfluidics
Laser photoablation	Laser photoablation is used to write directly onto a polymer layer with a high-powered laser, without photoresist or wet etching. The amount of material ejected can be tuned by adjusting the wavelength, energy density, and pulse width of the laser used for ablation.[30]	Flexible electronics; flexible displays
Offset printing	In offset printing, the inked image is transferred (or offset) from a blanket cylinder that bridges the plate cylinder and the substrate. The pattern is transferred to the blanket (usually made of rubber) and then transferred to the substrate.[28]	Print media
Inkjet printing	Inkjet printing is an additive technique involving the deposition of materials from one or more print heads. High-resolution inkjet printing often involves an array of piezoelectric print heads for the deposition of materials at precise locations. Lithographic patterning can further improve accuracy.[31]	Print media; displays; thin-film transistors; OLEDs

Figure 6.K.3 R2R Processing Diagrams for Organic Electronics/Thin Films[20]

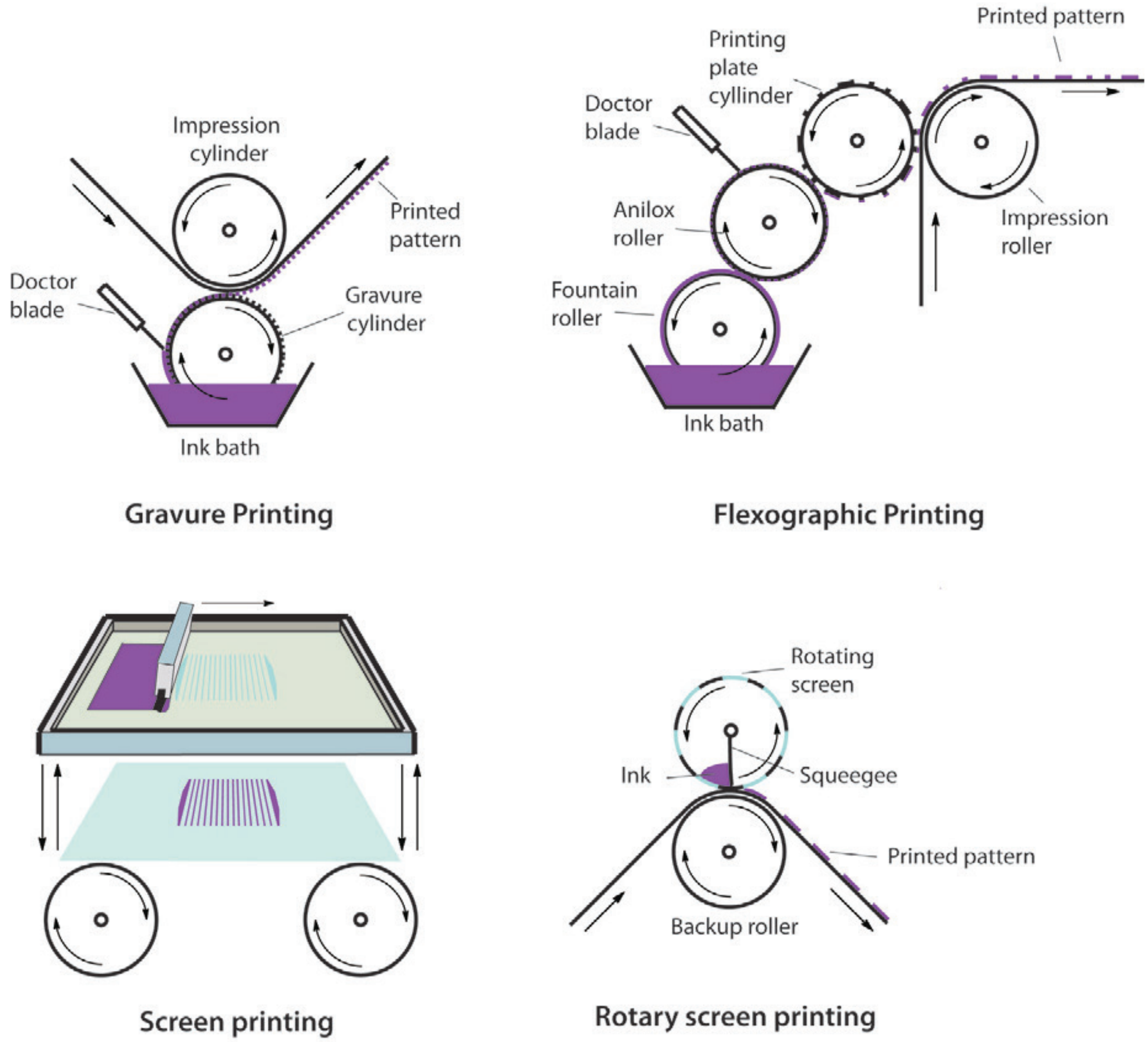


Table 6.K.4 provides a comparison of different printing methods and characteristics, impacting their theoretical capacity and practical applicability for large-scale R2R production.

Table 6.K.4 Comparison of Different Printing Methods and Characteristics Impacting their Theoretical Capacity and Practical Applicability for Large-scale R2R Production

Printing Method	Speed	Wet Thickness (μm)	Resolution (μm)	Start/Stop	Complexity	Applicability
Flatbed screen printing[32]	Low	5–100	100	Yes	Low	Limited
Rotary screen printing[32]	High	3–500	100	Yes*	Medium	Very good
Inkjet printing[32]	Medium	1–5	<50	Yes	High	Limited, materials must be jettable
Flexography[32]	Very high	1–10	<50	Yes*	Medium	Very good
SAIL[17]	High (>5 meters/min)		0.1 (100 nm demonstrated)			New technology
Laser photoablation[17]	Low		~10			Thermal effect sensitivity
Gravure [33]	High		>0.07 (70 nm demonstrated)			Very good

* Stopping should be avoided to minimize risk of registration lost and drying of ink in anilox cylinder. Short run-in length.

Technology R&D Needs

This section examines manufacturing R&D needs for R2R processing across the broad range of material sets and markets discussed previously.

Lithium-Ion Batteries

For lithium-ion battery electrode production, advancements in continuous materials deposition on webs to build the multilayer configuration by using tape cast, screen print, vapor or wet chemical deposition, or evaporative/sputter techniques are needed. Deposition of carbon nanotubes and whiskers on graphene are of interest for certain applications.[34] Additional needs include elimination of expensive solvents and recovery systems through aqueous processing, material processing cost, and electrode QC.[35]

Windows and Solid State Lighting Products

Applications of R2R products in buildings include solid state lighting products (e.g., LEDs and OLEDs) and dynamic window films. For solid state lighting products, high costs are a key barrier; new, lower-cost techniques are needed for materials deposition and device fabrication. Transformative processing techniques with the potential to significantly change the cost structure for these products—such as solution-processable coatings and novel encapsulation techniques—are of particular interest.[36] For R2R applications in windows, there is a need for improved coating processes, especially rapid, low-cost glazing coating processes that offer high yield, durability, and quality. Similar process needs are identified for the early-stage R&D topic of dynamic windows with energy harvesting, including manufacturing processes to achieve defect tolerance with minimal resistive losses and cost-effective, high-yield production at large scale.[37] For more information on these R&D needs, see *Supplemental Information for QTR Chapter 5: Building Energy Technology Roadmaps*.

Membrane Technologies

Areas of interest for membrane advancements include high pressure ceramic membranes, indoor air quality and dehumidification membranes for applications in buildings, water processing membranes, gas separations for natural gas processing and CO_2 capture applications (CO_2/N_2, CO_2, H_2, and CO_2/CH_4), liquid/gas separation membranes (CO_2 loaded solvents), forward osmosis capacitive polarization membranes, ionically conductive membranes, and other multilayer systems, such as those used in battery applications.[33, 38]

Photovoltaics

R&D needs related to R2R processing of OPV include the following:[39]

- Rapid processing and process control during manufacture.
- Processes that enable the printing of semitransparent electrodes.
- Processes that enable complete fabrication of efficient solar cells.
- Processes that enable thin outlines with low materials consumption and low embodied energy.
- Active materials and inks that leverage the thermo-mechanical properties of multilayer structures.
- Better control of film thickness, especially the uniformity of dry films in the multilayer stack through proper ink design. Film thickness control depends on the viscosity control, ink stability over time, ink rheology during deposition, ink rheology during drying (i.e., heating and up-concentration of solutes in the wet film), wetting behavior during deposition and drying, and control over morphology formation.
- Improved control of registration (the process of aligning the different layers) during R2R processing. Ultraprecise multilayer processing plays an important role for many types of devices, such as organic TFTs, especially for processing at very high speeds.
- Aqueous coating processes that enable true large-scale production. The use of different types of ink formulations (such as dissolved solutions, emulsions, and dispersed nanoparticles) as replacements to orthogonal solvents more typically used for the processing of multilayer devices could be useful.
- Replacement of toxic and chlorinated solvents typically used for processing of conjugated polymers with something less harmful.

Fuel Cells

The Fuel Cells Technology Office (FCTO) has cataloged needs related to R2R production of PEM MEA materials in the *Manufacturing R&D* chapter of the FCTO multiyear program plan.[40] These needs include further development of direct coatings of electrodes onto membranes and highly uniform lamination processes for MEA components as well as development needs for in-line QC of all MEA components. These focus areas are also highly relevant to PEM electrolysis cells. Registration of the anode and cathode coatings on PEM cells was also found to be of critical importance to mitigating degradation during operation.[41] The registration issue is further complicated for PEM cells because the two electrode coatings are not adjacent, as in many other applications, but are separated by the membrane.

Flexible Electronics

R&D needs for flexible electronics include transition from the plate-to-plate standard lithography typically used in industry to continuous and hybrid approaches to R2R processing and the development of advanced materials and equipment to enable commercialization of nano-imprint lithography and patterning with 50 to 100 nm print resolution at process rates of 3 to 5 meters per minute.[42]

For flexible "hybrid" electronics, which comprise flexible substrates and conductive elements integrated with traditional semiconductor chip architectures, several manufacturing challenges related to continuous processing include the following:[43]

- Low-temperature processing on bendable, stretchable, and/or foldable substrates, such as conformal coating on textiles and plastics
- Reliable, high-speed registration techniques for multilayer devices
- Finer features for higher input/output count die
- High-throughput and large-area printing/deposition systems that can handle a wide range of materials/inks and substrates with enhanced process control
- Deposition on nontraditional substrates (textiles, low-temperature plastics, stretchable materials, breathable materials) with varying surface energies, roughness, etc.
- Methods for depositing vias (small openings for an electrical connection) in multilayer circuit boards
- Precise registration for multilayer devices
- Understanding the interplay between functional materials, substrates, and deposition processes
- Developing multi-physics tools (electrical, thermal, mechanical, etc.) to determine device manufacturing layout
- Populating databases with both materials properties and fabrication process parameters

Cross-Cutting R&D

While many of the needs discussed above are specific to a particular material set or market, several needs are crosscutting, such as the need for precision registration of multiple coatings, aqueous ink development, and further development of multilayer coating techniques. Another general need that can have crosscutting implications is the development of atmospheric pressure alternatives to vacuum processes. In addition to the applications discussed above, this need has been identified for dynamic window films,[44] displays and optical films,[45] and thin film PVs.[46, 47]

An additional crosscutting need that is referenced in discussion of nearly every advanced and emerging product made with R2R processing is QC through metrology. Quality control is critically important for many, if not most, of the advanced clean energy and flexible electronics products discussed in this document that are or could be fabricated by using R2R processes. First, because R2R is a continuous process, in-line inspection of R2R processed materials is critical because of the cost, yield, and time risks in waiting until a roll of product is made to determine if the quality is acceptable. As noted by Gregg et al., R2R processing "provides a manufacturer with a means to rapidly increase capacity and productivity. However, the specific process in use, and all tools, materials, designs, etc., must be reliable, reproducible, and be capable of consistently yielding product to the prerequisite specifications."[2] Second, many clean energy products are highly functional and operate in extreme environments of temperature, humidity, pH, voltage, etc., such that even small defects in the layers can cause poor performance, hasten degradation mechanisms, and lead to premature failure.[48, 49, 50]

A recent DOE workshop[51] on QC identified a number of cross-cutting needs relevant to R2R processing including: thickness measurement; inspection for mechanical defects such as pinholes and cracks; measurement of electrical properties such as resistance; measurement of surface texture, structure and morphology; and inspection for interlayer delamination and voids. The workshop determined that research is needed to investigate these issues with respect to differences in scale, criticality, application, and ex situ measurement, while advancing tools and methods for the collection, analysis, storage, and use (either in real time or for later data mining) of high volumes of in-line QC data, and for the integration of these data into process control and feedback systems. Ultimately, development of models and methodologies to predict/correlate defects to performance would be a prime measureable program metric.[51]

Techniques currently used in industry to identify and quantify defects in materials include visual detection systems for cracks, fluorescence of functional coatings applied to textiles, noncontact eddy current measurements for surface sheet resistance, noncontact optical measurements for bandgap and relative thickness of coatings, noncontact XRF techniques to measure the composition and thickness of coatings, and photo-imaging for physical defects.[52, 53, 54] One limitation of current QC techniques is a lack of standards. In some cases, hardware is commercially available but the main gap is software relevant to specific applications. Systems are needed that can scan the composition of coatings (for multi-material coatings) and physical defects across the full width and length of the web while the web is in motion. Some measurements needed for in-line QC that current techniques do not address are physical defect density and/or pinhole density, bandgap measurements, surface sheet resistance of some coatings, optical transmission, relative thickness of coatings across and along the length of the web, and material composition measurements. Examples of challenges that need to be addressed by effective QC include the following:

- **For flexible electronics:** Defects can cause open and short circuits, leading to device failure. Metrology needs include discrete defect detection, surface roughness measurement, inspection of layer quality, measurement of electrical properties to ensure proper functionality, registration control, and possibility for repair/correction of defects.[55] In addition, inspection techniques are needed for pattern-specific defects, down to the 1 μm scale.[56] Several factors that can cause defects include missing or damaged/clogged nozzles in the print head, particles on the substrate, particles on the screen/stamp, web wander, non-uniform web tension, and mis-registration. Significant challenges remain for monitoring of high-throughput processes having nanoscale features. In combination with in-line inspection, model-based real-time diagnostics and control would complement the development of process modeling and control methods.[57] Realization of successful metrology and instrumentation for flexible electronic systems would open possibilities for the deployment of high performance flexible electronic components in a variety of applications, including communication, sensing, medicine, agriculture, energy, and lighting.[55] The Department of Defense (DOD) has also identified in-line, high-speed, automated QC tools—both for measurement of performance (e.g., electrical) and registration/geometry—as a key need for flexible hybrid electronics.[43]
- **For thick-film and thin-film microelectronics industries:** Substrate surface imperfections, chemical impurities, inconsistencies in substrate thickness, and inadequate surface flatness and planarity can all result in considerable yield loss. These losses can be compounded by other issues, such as improper deposition of conductors, devices, vias and other through-holes, mask alignments, etch issues, and oxidation. Pre- and post-fabrication yield losses during ingot preparation, doping, thermal processing, wafering, and dicing can also occur, including mechanical issues as simple as product breakage. Overall net yields for thick-film transducer and thermistor products can be as low as 25% to 30%, while those encountered for select discrete thin-film fabricated products are typically 40% to 60%.[2]
- **For thin film solar cells (TFSCs):** A critical area is maintaining process control and high yield. It is costly to fabricate a few-kilometer-long roll, or to make hundreds of square meters of glass TFSC modules, only to discover at final testing that a process-induced defect requires the entire batch to be scrapped. In-line process control and diagnostics have been applied to both $Cu(InGa)Se_2$ and amorphous Si based manufacturing. High yield is widely acknowledged as a critical issue for TFSC manufacturing.[58] Additional details on metrology and QC needs for both Si-based and thin film PV devices are provided in the DOE Solar Energy Technologies Office (SETO) *Metrology Workshop Report*.[59]
- **For scale-up of automotive fuel cell MEAs:** Debe[60] noted that a production rate of 11,700 MEAs/minute would be needed to produce 4.5 billion MEAs per year (10% of the world market in 2030, assuming 15 million fuel cell vehicles manufactured per year). To achieve a quality requirement of 0.1% stack failure, only one critical MEA failure in 300,000 would be allowed; for six sigma stack quality, only one critical MEA failure in ~90 million would be allowed.[60]

A final crosscutting need to further advance R2R processing, especially the ability of small- and medium-sized enterprises to validate new product constructions and manufacturability at an appropriate scale, is the need for pilot and/or shared development facilities. This infrastructure would provide a vital step between lab coupon-scale development and production line scale-up, ultimately reducing risk and cost and providing a more rapid development path for product commercialization. The challenges of high-cost R2R instruments could be alleviated by creating shared facilities accessible for academics and industrial participants to experiment with new ideas and establish emerging processes and materials for broader use.[57] Several for-profit entities with extensive history in the development of R2R processes have recently become active in this space, including Eastman Kodak,[61] Carestream,[62] and the Xerox Palo Alto Research Center.[63] Exploration of how these and similar companies might contribute to overcoming the R2R needs discussed herein in a more centralized way may be of interest.

Emerging R2R processes that exemplify these needs are described in the following sections.

Atomic Layer Deposition (ALD)

ALD is a thin film deposition technique that involves growing films through the sequential pulsing of chemical precursors onto a substrate. Precursors are introduced in sequence, separated by system purges, resulting in a uniform, pinhole-free, and conformal multilayer structure that can be adjusted by controlling the number of cycles.[14] ALD is suitable for depositing metal oxide films and certain metal coatings. Benefits of ALD include the relatively low-temperature processes and reasonably low-cost precursors needed for most applications. ALD techniques are being applied in the semiconductor industry and have been scaled to large-area substrate processes for thin film PVs and displays where the metal oxide coatings yield superior barrier coatings and dielectric films.[57] In addition, ALD is being pursued as a method to form extremely thin conformal coatings to improve the cycle life of lithium-ion cells.[35]

Potentiometric Stripping Analysis for Electroplated Alloys

Electroplating is an additive, solution-based deposition process and a suitable R2R platform for a range of metals and alloys. However, the key challenges for continuous, high-speed coating systems are the control of stoichiometry and the depletion of the plating baths in web-based systems. Potentiometric stripping analysis techniques precisely control both stoichiometry and uniformity of metal and alloy coatings on flexible webs. Other parameters for maintaining sufficient process control in the electroplating steps include keeping the solution at the work surface fresh and evenly biased by agitating the bath, providing adequate circulation, further using an inert environment such as an argon blanket to minimize the effects of oxidation, and using a separate anode for precise control of field distribution.[57]

High-Temperature R2R Processes

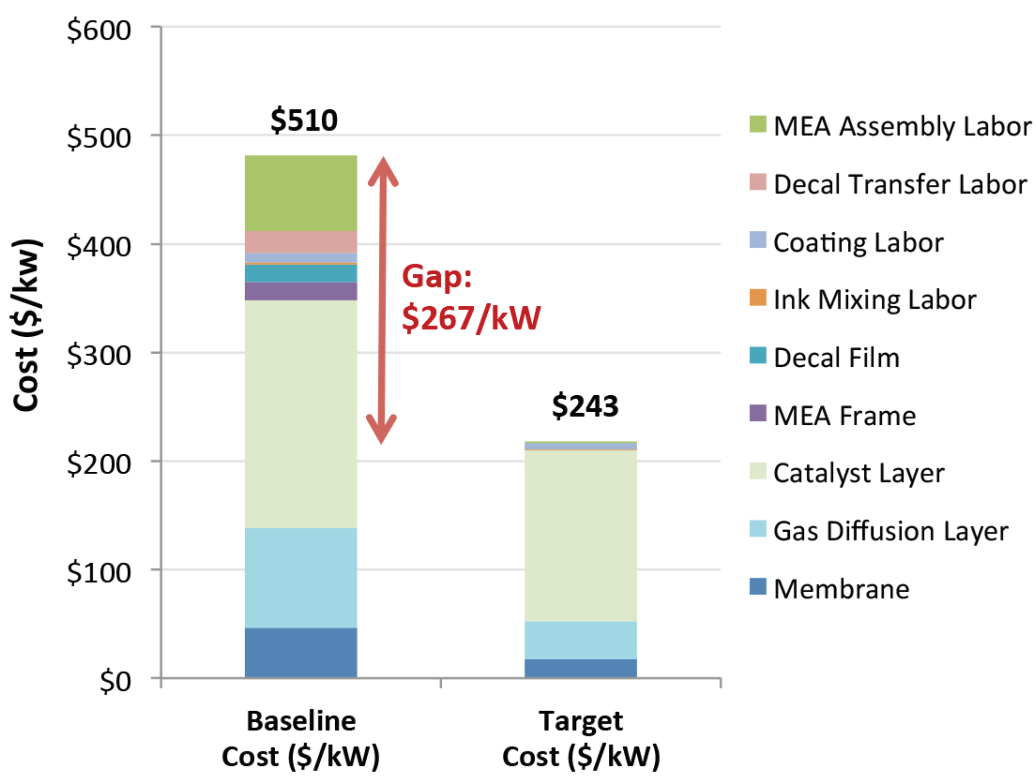
ORNL is conducting research in the development of high-temperature R2R processes suitable for creating crystalline, high-performance semiconducting materials. By using a high-temperature metal or other suitable substrate, it is possible to increase the high-temperature process capability to above 1200°C. The issue of interdiffusion may be resolved by depositing a stack of buffer layers to provide the required crystal orientation that includes deposition of a diffusion barrier and then active layer coating. The high-temperature R2R processes can be used to develop hybrid solution-based approaches as well. The high-temperature processes are suitable for a range of thin film crystalline materials, including silicon for solar PV, diamond, and other semiconductors. This R2R process capability opens up opportunities for large-area, high-quality semiconductors having electronic transport properties approaching those of bulk materials, thereby enabling high-performance electronic devices and systems.[57]

Potential for Cost Reduction

The potential for R2R to provide cost improvements is greatly dependent on the material set and market of interest. The following examples show how improvements in R2R processing can reduce the costs of new clean energy related products:

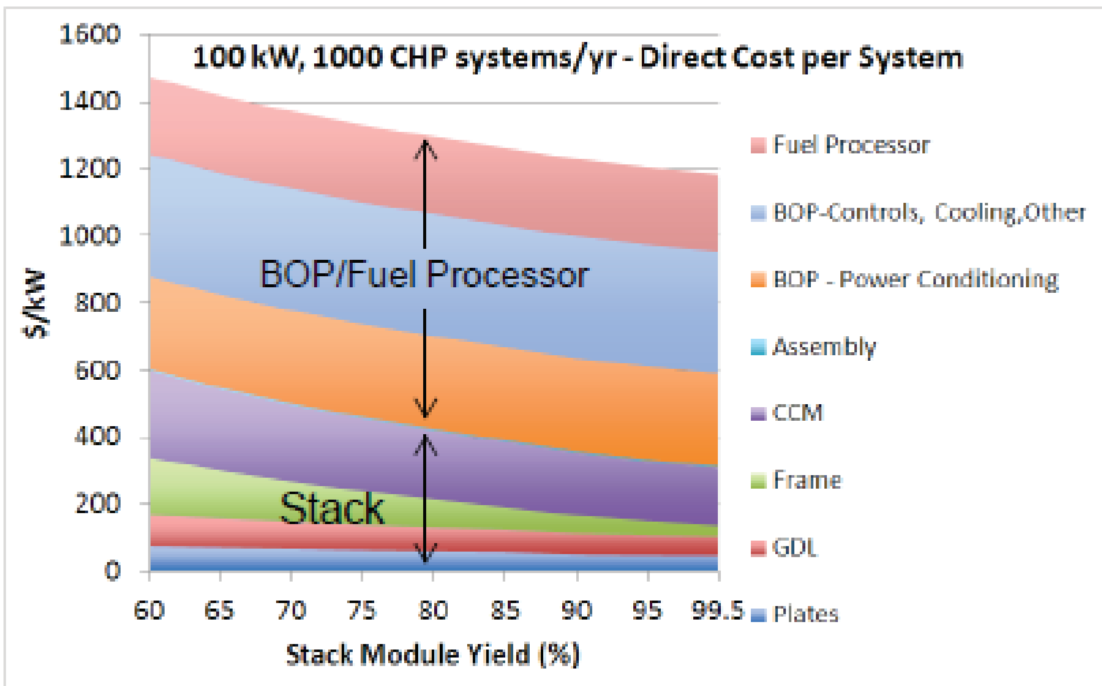
- Flexible printed circuit fabrication using batch processing and rigid-panel processes is expected to be superseded by less costly R2R technology to take advantage of high-speed plating, improved etching uniformity, intelligent material handling, and improved fluid delivery and dynamics.[64]
- R2R processing could enable low-cost production of fuel cells. An estimate of the R2R cost reduction potential for membranes, gas diffusion layers, and catalyst layer coatings for PEM fuel cells is shown in Figure 6.K.4.[65]

Figure 6.K.4 Strategy for Meeting Cost Targets for Automotive Proton Exchange Membrane (PEM) Fuel Cells through the Use of R2R Processing Techniques[65]



- Battery electrode production cost can be reduced by further advancements and improvements in R2R coating and processing. Aqueous electrode processing could potentially reduce the electrode processing cost and energy consumption by an order of magnitude. In addition, doubling the thickness of the electrodes could reduce the need for inactive current collectors and separators by 50%, therefore cutting the associated costs in half. These improvements, along with a reduction of the anode electrolyte wetting and solid electrolyte interface layer formation time, collectively offer the possibility of reducing lithium ion battery pack cost from $503/kWh-usable to $370/kWh-usable, a savings of $133/kWh (or 26%).[66]
- The potential cost impact (and by extension, potential savings) of poor QC resulting in low manufacturing yields is shown in the life-cycle cost analysis of PEM combined heat and power fuel cell systems (Figure 6.K.5). High levels of QC are needed during R2R processing of the cell materials.[67]

Figure 6.K.5 Fuel Cell System Cost as a Function of Stack Module Yield[67]



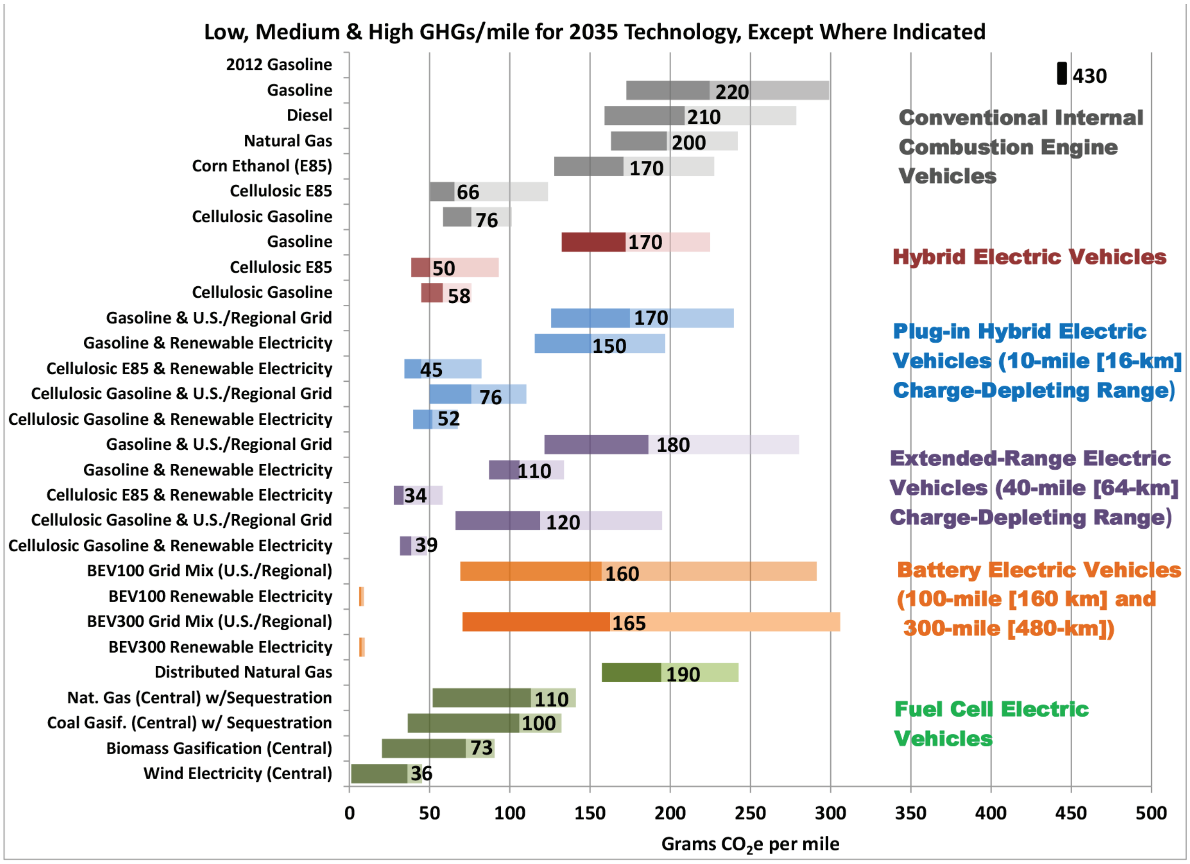
Potential Energy Impacts

The following examples show how improvements in R2R processing can improve energy utilization throughout the clean energy economy:

- According to a DOE/BTO analysis, today's state-of-the-art electrochromic windows have the potential to save approximately 1,200 TBtu and 720 TBtu from the residential and commercial sectors by 2030, respectively, if all the windows in new and existing buildings are replaced, regardless of the cost of the windows.[37]
- The replacement of traditional lighting systems with phosphorescent OLED lighting could result in cumulative U.S. energy savings of 220 TBtu and global carbon emissions reductions of 3.7 million metric tons between 2012 and 2018 (estimate made in 2011).[68] Konica Minolta, Inc., has constructed a plant to enable the mass production of flexible OLED lighting panels to expand its OLED lighting business beginning in fiscal year (FY) 2014.[69] U.S. companies such as Plextronics are forming partnerships to improve production of flexible OLED lighting and signage through R2R processing.[70]

- In the case of fuel cell and battery electric vehicles, the projected well-to-wheels greenhouse gas (GHG) reductions (Figure 6.K.6) are enabled, in part, by advances and scale-up of R2R processing of cell materials.[71]

Figure 6.K.6 Comparison of Well-to-wheels GHG Emissions of Battery and Fuel Cell Vehicles to Other Vehicles in the Fleet[71]



- Recently developed CO_2 capture membranes manufactured by using R2R processes, in combination with a novel process design that uses incoming combustion air as a sweep gas to generate driving force, show promise for meeting DOE CO_2 capture cost targets. Estimates indicate that this membrane process can capture 90% of CO_2 in flue gas as a sequestration-ready supercritical fluid using ~24% of plant energy, at a cost as low as $39 per ton of CO_2 captured (not including transportation to sequestration sites). Membrane permeance improvements or cost reductions may be able to further improve this process.[16, 72]

Program Considerations to Support R&D

Strategies and Pathways to Achieve Desired Impact

Technology Roadmaps Applicable to R2R Processing

R2R processes can make use of many different technologies for different applications. No specific technology roadmap exists for developing R2R processes in general; instead, roadmaps exist for specific technologies that would use an R2R process as the manufacturing method.

- The International Electronics Manufacturing Initiative (iNEMI) developed a technology roadmap for flexible electronics that addresses materials (nanoparticle suspensions, particle blends, and small molecular solutions), printing technologies (contact and noncontact), and processes (R2R, roll to sheet, and sheet to sheet).[73]
- The Centre for Process Innovation (UK) developed a technology roadmap to expand R2R and encapsulation processing technologies to target the development of flexible optoelectronic devices for the emerging printed electronics markets and to address many of the challenges encountered in scaling up emerging technologies to commercialization by adopting R2R processing techniques.[74]
- From an industry perspective, Baker™ Wet Process Equipment has developed a technology roadmap to understand and manage the issues associated with conventional versus R2R processing for manufacturing flexible printed circuits.[64] Their roadmap focuses on the core of current manufacturing trends toward producing thinner, lighter, and higher density printed circuits by use of effective handling and processing of a thin core material. R2R processing equipment will need to focus on smooth, steady transport of films through various wet processes in both a horizontal and vertical plane, and will require next generation spray- or immersion-type technologies.
- The National Aeronautics and Space Administration (NASA) has drafted integrated technology roadmaps for 15 Technology Areas that include "pull" and "push" technology strategies and consider a wide range of pathways to advance their current capabilities in space. Technology Area 12 addresses materials, structures, mechanical systems, and manufacturing. Although R2R is not specifically addressed as part of the NASA roadmaps, several of the technologies and processes—such as hybrid laminates, polymer matrix composites, multifunctional thin films, flexible materials for entry-descent-landing, PVs, lightweight aluminized thin film systems for solar sails, and large ultralight precision optical materials—are all directly applicable to R2R processing.[75]

Workshops on R2R Processes and Manufacturing

Workshops can enable the capture, integration, and cross-pollination of the knowledge of a broad range of experts from industry, universities, national laboratories, non-profits, and the public sector, and can help identify critical opportunities and pathways for R&D, and build broad expert community engagement.

In 2015, DOE AMO hosted a workshop on high value R2R technologies.[76] Participants in the workshop identified the following cross-cutting challenges and barriers impacting R2R:

- **Lack of modeling tools:** Improved tools are needed to accurately model R2R deposition processes and provide insight on alternative materials and process routes.
- **Scalability not demonstrated:** There is a lack of established infrastructure and processes to demonstrate R2R processes at practical line speeds.
- **Inadequate materials and substrates:** Materials and substrates are needed to achieve unique performance features such as rapid drying and transparency.
- **Insufficient defect control and monitoring:** Real-time, in-line quality monitoring and control capabilities are needed to enable defect identification and process parameter adjustment.
- **Lack of collaboration:** Accessible demonstration and testing facilities, key for mitigating upfront risk for new and smaller companies entering the market, are not available.

For technology areas impacted by R2R processing, workshops such as the National Institute of Standards and Technology (NIST)/National Nanomanufacturing Network Workshop on Nanofabrication Technologies for Roll-to Roll Processing[57] and the DOD/DOE Manufacturing Innovation Topics Workshop have provided valuable information.[77] Discussions at these workshops have focused on using R2R processes for coating of polymer films and device level patterning, and nanoimprint lithography (NIL) for R2R processing of nanotechnologies.[57] Workshops also address programmatic issues for R2R processing, such as R2R process technology needs, manufacturing challenges and investments, process deficiencies and metrological needs, and quality control systems.[37, 59, 77, 78] Nanomanufacturing workshops are held annually to provide opportunities to share information on emerging processes and scaled manufacturing platforms where R2R may have a role.[57, 79] They also focus on specific technology areas that have immediate applications to clean energy initiatives, such as biomass indirect liquefaction that focuses on pathways that convert biomass-based synthetic gases to liquid intermediates.[80] Common areas of interest lie in overall technology needs, manufacturing challenges, and investment levels.

Enabling Science Activities

Given the breadth of materials, processes, and needs encompassed by R2R processing, enabling science opportunities are broad. Potential key areas of enabling science to address the needs previously discussed in the *Technology Assessment and Potential* section include the following:

- Rapid, high resolution imaging
- Interface and surface science
- Process modeling
- Rheology and colloid science
- High-speed data acquisition and processing

Crosscutting Technologies and Issues

Most R2R process technologies are crosscutting and can be used for many different material sets, often over a wide range of operating conditions and for products suited to many different markets. For example, further advancement of dual slot die coating would be beneficial for both OPV films and electrochemical electrodes, among many other applications. And as discussed earlier, QC is clearly a crosscutting issue. Efforts to advance these and other R2R technologies or technology enablers could produce enduring economic benefits for both the public and commercial sectors.

Partnerships and Stakeholder Engagement

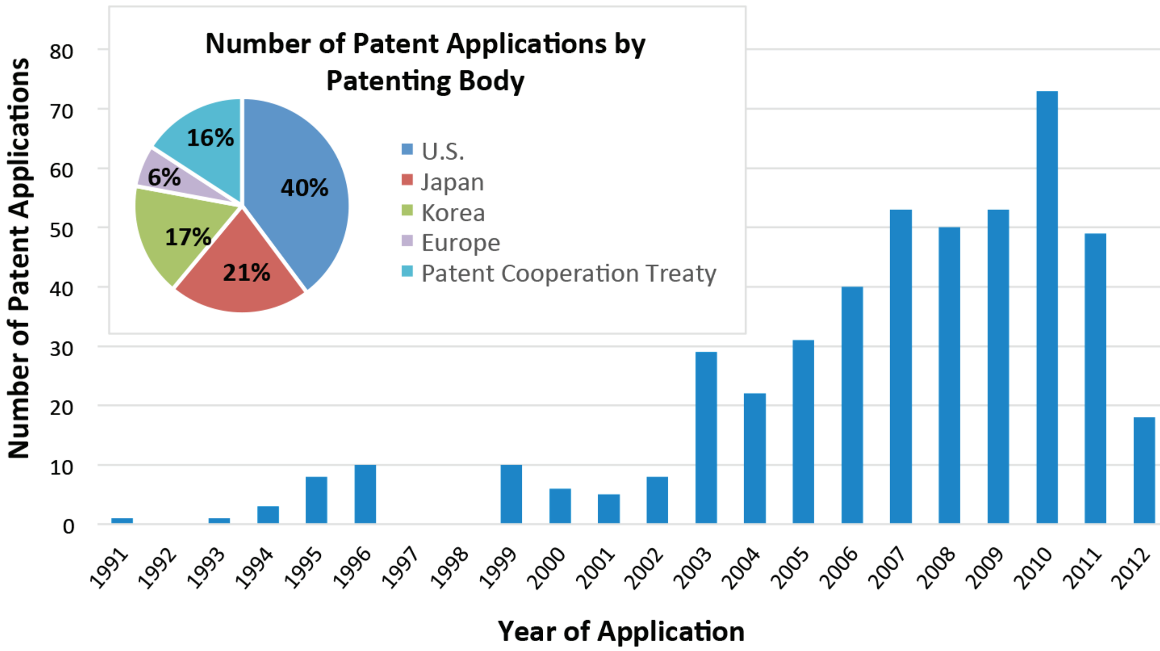
Investment in R2R processing is increasing in different market segments across a variety of applications, including investments in energy saving technologies that can be produced by R2R processing processes. Examples of organizations actively developing and applying these methods include the following:

- **Companies:** Large companies include 3M, DuPont, General Electric, Dow, Avery Dennison, Corning, HP, Gore, Merck, Sumitomo, Kateeva, Tokyo Electron, and BASF.
- **Industry groups:** Developed to support clusters of companies in certain markets, including the FlexTech Alliance, AIMCAL (for metallized films), and the Paper, Film & Foil Converters Association.
- **Government:** DOE and DOD support ongoing activities in their areas of interest. The National Science Foundation (NSF) supports basic research related to the materials, physics, characterization, and instrumentation that are applicable to R2R processes.
- **National labs:** Lawrence Berkeley National Laboratory (LBNL), ORNL, National Renewable Energy Laboratory (NREL), Pacific Northwest National Laboratory (PNNL), and others are pursuing work in this area.
- **Academia:** University of Massachusetts—Amherst, University of Louisville, Binghamton University, University of Minnesota, University of Texas at Austin, Arizona State University (ASU), and others are pursuing work in this area.

- **International:** The Fraunhofer Institutes in Germany, amongst many other international organizations (public and private), are engaged in R2R development.

A 2013 report by Information Handling Services evaluated 483 R2R processing technology patents related to flexible electronics. As shown in Figure 6.K.7, the number of these patent applications has steadily grown. Major applicants in the United States include 3M Innovative Properties, SiPix Imaging, Fuji Film, and General Electric.[81]

Figure 6.K.7 R2R Processing Technology Patent Application Trends by Year[81]



Current Research Efforts by DOD

In August 2015, DOD awarded a Manufacturing Innovation Institute (MII) for Flexible Hybrid Electronics to a consortium of 162 companies, universities, and nonprofits led by the FlexTech Alliance, located in San Jose, California.[82] This is the ninth MII announced to date by a U.S. government agency.[83] In total, the Flexible Hybrid Electronics MII will receive $171 million to invest in strengthening U.S. manufacturing ($75 million in funding over five years, matched with more than $90 million from industry, academia, and local governments).[84]

The Air Force Research Laboratory (AFRL), the Army Research Laboratory (ARL), and the Naval Research Laboratory have all been actively cooperating on micro-electronics research focused on flat panel displays as follows.[85]

- ARL has sponsored research in thin film transistor arrays for displays and digital X-ray detectors. Currently, ARL manages the Flexible Display Center (FDC), based out of Arizona State University (ASU). The FDC is a unique public–private partnership with the goal to accelerate the availability of flexible display technology for soldiers. Some results of FDC work include the demonstration of the world's first flexible electrophoretic display (E-ink Corporation), using the ASU-patented bond–debond manufacturing process; production of the ultra-large format flexible full color OLED displays (14.7-inch diagonal) and X-ray detector arrays (FDC—Defense Threat Reduction Agency [DTRA] and the Palo Alto Research Center [PARC]); development of a range of handheld devices with an integrated flexible reflective display (E-ink Corporation); demonstration of flexible reflective displays and fully flexible tablets used in Army field experimentations (Physical Optics Corporation); and development of flexible microelectromechanical systems among others.[86]

- Starting in FY2011, DTRA and ARL are collaborating to develop flexible digital X-ray detectors; while the manufacturing process is currently plate-to-plate lithography, R2R is under consideration.
- A project with Hewlett-Packard and PowerFilms was designed to advance a plate-to-plate and R2R SAIL process for display applications on the basis of amorphous silicon (Si) TFT arrays. Although the program was concluded without commercialization of the technology in FY2011, process feasibility was demonstrated. Efforts are now continuing via an ARL and FlexTech alliance focused on SAIL development.
- The ARL has made investments through the FlexTech Alliance, a 30 industrial member consortium of both domestic and international organizations. Their focus includes work on zinc-polymer battery chemistries (referred to as Imprint Energy) that can be processed by using screen printing fabrication approaches. The effort also included TFTs and R2R processed OLEDs, among others. The FlexTech Alliance also sponsored flexible Si complementary metal-oxide-semiconductor (CMOS) chips on paper, soldier health monitoring systems, and other electronics designed to provide and enable prognostics and diagnostics. Many of these exploratory programs are at a technology readiness level (TRL)[87] of 1–3. The paper-based flexible Si project is at a TRL of 6 and a manufacturing readiness level of 3–4. This project represents a nontraditional flexible Electronic Manufacturing Services program.

Current Research Efforts by DOE

DOE supports R&D in the area of fuel cells, energy efficient buildings, solar energy, batteries and electric vehicles, advanced manufacturing technologies, and fossil fuel energy as part of a broad portfolio of activities to secure the nation's energy future. As an example, the DOE 2014 Manufacturing Roadmap for Solid State Lighting Research and Development[88] developed with extensive input from industry, universities, national laboratories, and others lists luminaire manufacturing, test and inspection equipment, and phosphor manufacturing and applications as the top three priorities for LED manufacturing R&D as well as OLED fabrication equipment, OLED substrate and encapsulation manufacturing, and OLED panel manufacturing as the top three priorities for OLED manufacturing R&D. All of these involve some form of R2R processing.[89] DOE technology offices' efforts include the following:

- Fuel Cell Technologies Office (FCTO) supports process development and development of in-line QC techniques for R2R proton exchange membrane fuel cell MEA production.[40]
- Solar Energy Technologies Office (SETO) (through the SunShot Initiative) invested $30 million (with 50% cost share matching from industry and other partners) to establish a consortium called the U.S. Photovoltaic Manufacturing Consortium (PVMC) in Albany, New York, to support CIGS PV products. SETO has recently redirected the consortium to work on "downstream" issues in support of flexible CIGS, such as establishing methods for installation of flexible PV modules and assessing flexible PV reliability.
- Building Technologies Office (BTO) has a number of existing investments in R2R processing, including research on:
    - Airflow panel membranes for architectural applications (LBNL);
    - R2R sensors for building applications (ORNL);
    - Vacuum insulation (VI) window films (NREL);
    - Low-energy/electrochromic window films (ITN Energy Systems and the Electric Power Research Institute);
    - Daylighting films for windows (3M and LBNL);

- Primer-less, self-adhered air sealing membranes (3M and ORNL/China Clean Energy Research Center program);
- Infrared-responsive window coatings (PPG Industries and PNNL);
- Near-infrared electrochromic window coatings (Heliotrope Technologies).

- Vehicles Technologies Office (VTO) has projects exploring R2R methodologies for improvements in battery electrodes at ORNL and NREL.[35]
- Advanced Manufacturing Office (AMO), through prior programs supporting inventions and innovation as well as industrial sensors and Small Business Innovation Research (SBIR), has conducted competitive solicitations for cost-shared activities and grants resulting in the investment of approximately $1 million in CdTe solar cell development and manufacturing; approximately $1 million in advanced solar-reactive glazing, coating, and manufacturing technologies to reduce unwanted solar gain through windows, skylights, and automotive windows; approximately $1 million among battery technologies, super-capacitor technologies, and superconducting cable technologies; and approximately $2M in advanced sensor technologies. These AMO investments focused on lithium-ion battery technology incorporating R2R processing. A Manufacturing Demonstration Facility (MDF) has been established at ORNL, with a focus on electrolyte materials used in laminated planar battery pack assemblies.
- Office of Fossil Energy (FE) has made investments in CO_2 membranes. R2R processes are being considered or used to manufacture several different polymeric and ceramic/metallic membranes for CO_2 separation for power plants. Similar processes are used to manufacture existing commercial water filtration and natural gas processing membranes. Many of the technologies are at the pilot scale, and many of the manufacturing processes efforts are considered to be at a similar scale of development (TRL 4–5). Investments in membranes made to date, detailed in Table 6.K.5 for post-market and pre-market applications, may benefit from a concerted effort to improve the R2R processing processes.[38]
- NREL is addressing QC needs for scale-up of PVs, fuel cells, and battery electrode component manufacturing on continuous processing lines. The approach includes understanding QC needs from industry partners and forums, developing diagnostics, using modeling to guide development, using in situ testing to understand the effects of defects, validating diagnostics in-line, and transferring technology to industry.
- NSF supports fundamental and translational research efforts within the Center for Hierarchical Manufacturing, a Nanoscale Science and Engineering Center, leveraging $4 million/year of federally funded nanomanufacturing research. The research program focuses on the integration of nanofabrication processes for <30 nm elements on the basis of directed self-assembly, additive-driven assembly, nanoimprint lithography (NIL), high fidelity 3D polymer template replication, and conformal deposition at the nanoscale with silicon wafer technologies or high-rate R2R-based production tools. Within the Center for Hierarchical Manufacturing, the University of Massachusetts (Amherst) sponsors the "Research Cluster R: Roll-to-Roll Process Research Facility." The facility supports efforts focused on NIL process and development. Current capabilities allow work up to 6-inch wide format, using a range of R2R equipment and analytical tools. Focus areas include planarization, imprint embossing and patterning, alternative materials and membranes, functional hybrids, viscoelastic fluids, R2R integration, and design for manufacturability.

  NSF also supports fundamental and translational research efforts within the Nanomanufacturing Systems for Mobile Computing and Mobile Energy Technologies, an Engineering Research Center leveraging $4 million/year of federally funded research on innovative nanomanufacturing, nanosculpting, and nanometrology systems that could lead to versatile methods for the high-volume nanomanufacturing of mobile computing devices, such as wearable sensors, foldable laptops, and flexible batteries.

Table 6.K.5 Office of Fossil Energy Investments in CO_2 Membranes

Technology Stage	Company/Agency	Substrate	Active Layer	Type
Post-market	Ohio State University	Polymer (polyethersulfone)	Zeolites	Spiral Wound
Post-market	Membrane Technology & Research, Inc.	Polymer	Polymer	Spiral Wound
Post-market	General Electric	Polymer	Phosphazene	Hollow Fiber
Post-market	Gas Technology Institute	Polymer (polyether ether ketone [PEEK])	Perfluoro-oligimer	Hollow Fiber - Gas/Liquid
Post-market	Argonne National Laboratory	Metal Oxide (alumina-zirconia)	Pd/TZ-3Y Cermet	Long-tubes
Post-market	Pacific Northwest National Laboratory	Ceramic/Metallic	Ionic Liquid	Sheet/Plate
Pre-Market	Praxair	Ceramic	Pd Alloy	Shell and Tube
Pre-Market	Eltron	Metal Alloy	Not Applicable	Shell and Tube
Pre-Market	Worcester Polytechnic Institute	Metal Alloy (PSS-316L)	Pd Alloy	Shell and Tube
Pre-Market	Pall Corporation	Metal (zirconia-coated stainless steel tubes)	Pd Alloy	Shell and Tube
Pre-Market	Los Alamos National Laboratory	Polymer (polybenzimidazole [PBI])	PBI Polymer	Hollow Fiber

- The University of Kentucky Center for Applied Energy Research has a significant effort underway that focuses on a range of energy applications, some of which involve R2R. Areas of interest include low-cost carbon anode precursors, vanadium redox flow batteries, and thermoelectrics.
- The Center for Advanced Microelectronics Manufacturing (CAMM)—a partnership between Binghamton University, Endicott Interconnect Technologies, Cornell University and the Flex Tech Alliance—is a prototype R&D facility in large area flexible electronics. CAMM is part of Binghamton University's New York State Center of Excellence in Small Scale Systems Integration and Packaging, which serves as an international resource for systems integration and packaging R&D.

In addition to government and university activities, several industry groups are prevalent in different R2R markets and actively support member companies. Examples include the FlexTech Alliance, AIMCAL, and the Paper, Film & Foil Converters Association.

International Cooperation/Competition

Over the last 10 years, the European Union has made significant investments in R2R processing and related plate-to-plate printing of organic-based TFTs for displays and RFIDs. Organizations involved include Plastic Logic (focused on plate-to-plate), POLYIC (involved in R2R RFID), and Philips. Plastic Logic, in partnership with a supplier of OLED materials called Novaled, recently demonstrated a range of flexible electronic OLED displays designed as "wearable" devices.[90] Significant R2R processing capabilities also exist at several European laboratory organizations, including Fraunhofer COMEDD,[91] CEA Liten (France),[92] and Julich Laboratories (Germany).[93] Areas of study include fuel cells, OLEDs and other flexible electronics, and OPVs. In addition, PHOTOSENS, a European consortium led by VTT Technical Research Centre of Finland, is developing polymer-based nanophotonic sensors for air quality, pharmaceutical process cleanliness, and food safety applications.[94]

Risk and Uncertainty, and other Considerations

Market/Volume/Cost Challenges

In a continuous manufacturing process, the cost per unit length or area of processed materials decreases with product volume, due to economies of scale. R2R lines and process equipment can be very expensive to construct. They are not suitable for low volume products[95] unless the product can command a high margin. Large companies with extensive legacy investments in R2R equipment and capabilities have typically invested in smaller-scale, often very flexible pilot lines for smaller volume research or market development efforts. However, new entrants or smaller companies typically do not have access to these facilities and thus find process development and scale-up efforts challenging. Providing capabilities or facilities to facilitate the transition from emerging process R&D to scaled manufacturing would be of high value.[57] Similarly, in cases where other legacy processes are established with an industry standard approach, such as plate-to-plate processing of flexible electronics, exploring the transition to R2R can be difficult and, without shared facilities, extremely costly.[96] R2R production lines, especially the larger lines, are limited to the breadth of products that can be produced because of the large investment in a defined set and sequence of processes.[26] Modularity is somewhat difficult with R2R processing because the continuous motion of the substrate from one process to the next typically requires extremely close alignment of the rollers along the length of the line. However, modularity is possible, especially with smaller scale systems.

Technical Challenges

Because R2R processes run continuously, they inherently require a large quantity of feed materials. Thus new product (or process) development can be hampered by losses resulting from incoming materials variations, lack of process tolerances, and lot-to-lot variations. Newly developed or improved standards would assist the translation of discrete processes to an integrated manufacturing flow.[57] Process scale-up can be highly challenging (e.g., scaling from a 4-inch or 6-inch pilot coater to product widths of several feet or more while maintaining process uniformity). Transitioning from proof of concept with discrete substrates to a continuous process involves larger equipment and considerably different physics. Detailed investigations for R2R processing development could include tools to feed precursor solutions and slurries at sufficient rates, while controlling the rheology of these materials; development of substrates for improved tensile strengths, surface finishes, coating release, defect densities, etc.; development of motors and motor controls (web speed control, tensioning, material "take-up," post formatting, etc.); simulation and design tools; control(s) for feedback and adjustment; materials drying, curing, and heat treating accessories; ventilation and effluent treatments; incorporation of concurrent/simultaneous process by using additive and subtractive techniques; development of atmospheric and vacuum processing, precision alignment, and registration; and lithographic imaging and etch/deposition. Finally, for almost all new products, in-line metrology and inspection is a challenge because new products have new functional requirements and specifications, which are often not addressed (or not addressed well) by existing QC techniques.

Risks and Uncertainties of Using R2R Processes and Manufacturing

R2R processing is not ideal for every material and application. It is a promising innovation for high volume, large area production of thin and thick film materials with minimal defects and waste. R2R processes, in general, are energy efficient and environmentally friendly. However, as with any type of manufacturing, there are associated risks and uncertainties as follows:

- **High startup costs:** A combination of high costs, poor availability of production tools, and limited critical manufacturing technology developments are hindering the adoption of R2R processing for flexible electronics. For OLEDs, the startup costs for an R2R processing line are not clear, given that R2R production cannot match the performance of vacuum deposited products (OLEDs) at present, and some of the required layers must be vacuum deposited. Research is ongoing to develop R2R approaches for these layers and to improve R2R OLED performance levels. CAMM (in Binghamton, New York) is expanding its tooling capability to actively research R2R processing for emerging technologies such as large-area OLED lighting, PV cells on plastic substrates, low-cost RFID tags, and lightweight electronics and packaging platforms on rugged, flexible substrates. Further research in specific applications that employ R2R processes will provide the data needed to reduce the costs of startup. While there may be some commonality in the production technology, there will most likely be significant differences in the specific layers for different applications that preclude a general approach or analysis. As with many technologies, R2R faces the chicken-and-egg problem of needing a large market to develop and invest in production technology to get economies of scale and learning combined with an inability to develop such a market without the low costs enabled by large-volume production. If new product constructions are similar to existing products, however, an important strategy may be to use existing pilot or production lines until the new market supports a dedicated R2R production line.
- **Speed of high volume/large area process versus low volume/discrete process:** The speed and capacity for R2R processing versus a batch process is dependent on the material requirements for the end product and is directly related to costs. Until technology developments stabilize and application requirements are better understood for R2R products, it is difficult to know the extent of the trade-offs of different manufacturing approaches. The differences among applications will result in important differences in the manufacturing process for the different devices. Research is underway to improve manufacturing processes for both high volume/large area production as well as lower volume/discrete processes. Ultimately, determination of which approach is superior will depend on the requirements of the application as well as developments in flexible substrate materials, device structures, device materials, and encapsulation. At this time, issues of manufacturing scale are dependent on the application market, technical requirements, and performance trade-offs with the different manufacturing approach. The cost impacts of at-scale R2R processing can be mitigated in appropriate cases by utilizing already capitalized equipment developed for similar products or at shared facilities.
- **Material variations, tolerances, lot variations, and scrap:** Variations in substrates and products in an R2R production lot can be caused by several factors, including the materials used, the machinery involved, the control of the web, and the processes employed (lithography, deposition, etc.). Even the configuration of the rollers (double side mounted or cantilevered) can produce variations. In some applications, such as thin films and nanomaterials, the tolerances must be closely controlled in order to get a quality end product. If tolerances and variations are significant, then the R2R process can result in large quantities of scrap and waste material that may not be recyclable (and can also add to manufacturing costs). As the R2R process is adjusted in response to anomalies in the initial production phases, the material and lot variations are usually reduced, and the end products are brought within tolerances. Research is needed on various types of instruments that can be incorporated into the R2R process to further reduce variations and eliminate scrap. As above, this risk can be mitigated by the establishment and support of shared facilities.

- **Metrology:** As previously discussed, the success of employing an R2R process in manufacturing a specific technology is heavily dependent on process and cost control. Metrology tools compatible with R2R line speeds are needed to address defects (such as pinholes, nonuniform thickness, and impurities from static buildup), substrate quality, and registration (pattern position).[55] Proven techniques for process control include optical techniques such as transmission, reflection, and dark field imaging of printed or coated films, which can reveal film thickness variations, registration, and particle detection. These techniques are noncontact techniques and can be applied to individual layers during manufacture. Methods such as light-beam-induced current mapping, dark lock-in thermographic imaging, electroluminescence imaging, and photoluminescence imaging, are being used successfully today in R2R processing of solar cell materials.[20]

  Ultimately, in-line R2R instrumentation tools are needed that will test final product functionality, not just the presence of defects—for example, to assess the functionality of a solar cell itself (i.e., the production of electrical energy when subjected to illumination). Current in-line monitoring techniques are useful for guiding the process, but they cannot guarantee final device performance.

- **Proprietary information and intellectual property:** Successful implementation of R2R processes within the manufacturing industry will benefit from information exchange, resource partnering, and open discussion of ideas, discoveries, and best practices. Key challenges exist in providing an open forum for networking while protecting proprietary information and intellectual property.

Case Studies

High-Efficiency Thin-Film Solar Cells

Figure 6.K.8 Large-scale Thin Film PV Solar Power Plant[97]

Credit: First Solar

Working with DOE NREL and other partners, the U.S.-based company First Solar, Inc., recently achieved a new world record for CdTe PV solar conversion efficiencies, achieving 21.5% conversion efficiency in a laboratory cell and 18.6% conversion efficiency in a module.[98, 99, 100] Since 2011, greater improvements in CdTe PV research cell performance have been demonstrated compared to conventional multicrystalline silicon technologies, which have not changed appreciably for several years (current research cell world record is 20.8% by Trina Solar; see also Figure 4.P.5 in QTR Technology Assessment 4.P Solar Energy).[98] A promising feature of the CdTe cell is that it was constructed by using processes such as R2R and materials designed for commercial-scale manufacturing, potentially making it easier for First Solar to quickly scale up production. The advanced technologies and processes developed for the CdTe PV solar cell are already being commercialized and are expected to positively impact performance of future production solar cell modules and power plants (Figure 6.K.8).

Commercial Buildings Integration of Energy-Saving Window Coatings

Figure 6.K.9 The Energy Systems Integration Facility at NREL, which features dynamic electrochromic windows.[101]

The DOE Building Technologies Office (BTO) works with the commercial building industry to accelerate the uptake of energy efficiency technologies and techniques in both existing and new commercial buildings. BTO has several projects in R&D for development of electrochromic windows, high-insulating windows, and nano-lens window coatings for daylighting and low-emissivity storm windows.[102] Many of these products are fabricated with R2R processing technique, which is often the only practical and cost-effective manufacturing method. Examples of commercial energy-saving window products include 3M™ daylight-redirecting window films, which block up to 60% of the sun's heat without sacrificing daylighting or views;[103] SageGlass® electrochromic glazings, which dynamically adjust to control solar glare and heat gain;[104] and View Dynamic Glass, which also dynamically adjusts tinting to reduce glare and solar heat gain.[105] Electrochromic windows were recently installed at the state-of-the-art NREL Energy Systems Integration Facility, as shown in Figure 6.K.9.

Electrochromic window coatings are typically fabricated on non-flexible glass substrates, leading to the current high costs of these technologies. However, research is underway to deposit electrochromic films on low-cost flexible substrates by using R2R processing methods. R2R technologies could drive down costs and increase manufacturing throughput for these energy-efficient products. Also, flexible devices could enable retrofitting to existing windows.[106]

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Acronyms

ALD	Atomic layer deposition
CCM	Catalyst coated membrane
CIGS	Cadmium indium gallium selenide
CVD	Chemical vapor deposition
GDL	Gas diffusion layer
GHG	Greenhouse gas
LED	Light emitting diode
MEA	Membrane electrode assembly
NREL	National Renewable Energy Laboratory
OLED	Organic light emitting diode
OPV	Organic photovoltaic
ORNL	Oak Ridge National Laboratory
PEM	Proton exchange membrane
PV	Photovoltaic
R2R	Roll-to-Roll
SAIL	Self-aligned imprint lithography
TFT	Thin film transistor
TFSC	Thin film solar cell
UV	Ultraviolet

Glossary

Atomic Layer Deposition (ALD)	A thin film deposition technique that involves growing films through the sequential pulsing of chemical precursors onto a substrate.
Flatbed screen printing	A R2R-compatible printing process involving the use of a mesh stencil or "screen" to apply ink in the desired pattern. A squeegee is used to force ink through the screen and onto the substrate.
Flexible electronics	Electronic components produced on flexible or stretchable substrates. Flexible devices can be attached to curved and irregular surfaces such as buildings, objects, and human skin. Advanced manufacturing processes such as R2R can preserve the full capabilities of traditional electronics in conformable architectures such as these.
Flexographic printing	A R2R printing process that uses a flexible relief plate to transfer an image to a flexible substrate. During printing, the substrate material is fed between the inked printing plate and an impression cylinder, transferring the image as the substrate passes through. Flexographic printing is essentially a modern version of a letterpress.
Gravure	A R2R printing process that involves engraving an image onto a cylindrical image carrier. The patterned cylinder is covered with ink, then excess ink is removed with a doctor blade, leaving ink only within the recessed motif areas. Ink is then transferred from the image carrier to the substrate via capillary action.
Imprint or soft lithography	Any of several related methods for fabricating structures using elastomeric (soft) stamps or masks.
Inkjet printing	An additive R2R manufacturing process involving the deposition of materials from one or more print heads. High-resolution inkjet printing often involves an array of piezoelectric print heads for the deposition of materials at precise locations.
Laser photoablation	A R2R process used to write directly onto a polymer layer with a high-powered laser, removing material without photoresist or wet etching. The amount of material ejected can be tuned by adjusting the operation parameters (e.g., wavelength, energy density, and pulse width) of the laser used for ablation.
Light-emitting diode (LED)	A light source that relies on an electroluminescent semiconductor material to emit light when an electric current is applied.
Photovoltaic (PV) cell	A device that converts energy from light (e.g., solar energy) to electricity using semiconductor materials that exhibit the photovoltaic effect.
Offset printing	A R2R process where an inked image is transferred (or offset) from a plate to a blanket cylinder (usually made of rubber), and then to the substrate.
Organic light-emitting diode (OLED)	A light-emitting diode that uses an organic compound as its electroluminescent layer to emit light.

Organic photovoltaic (OPV) cell A solar cell produced using semiconducting organic materials.

Roll-to-roll (R2R) processing A low-cost, high throughput technique for continuous two-dimensional deposition of materials over large areas onto moving webs, carriers, or other substrates. Also known as web processing or reel-to-reel processing, R2R can be used to fabricate products on substrate materials including flexible plastic, glass, ceramic, composite, and metal foil.

Rotary screen printing A R2R process where a substrate moves through rollers past a squeegee, forcing ink through a cylindrical design screen onto the substrate.

Self-aligned imprint lithography (SAIL) An R2R fabrication method developed by Hewlett-Packard and PowerFilm Solar that involves imprinting a substrate using a single, three-dimensional masking structure. Because all pattern layers are incorporated into the mask, no sample alignment is necessary.

Vacuum deposition Any of several related methods for depositing layers of material onto a solid surface in a vacuum environment (i.e., at a pressure well below atmospheric pressure). Examples of vacuum deposition processes that can be successfully implemented in R2R processing include evaporation, sputtering, and chemical vapor deposition (CVD). Multilayer sputtering systems are the most common.