Review

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Recent advances in continuous nanomanufacturing: focus on machine learning-driven process control

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Abstract: High-throughput and cost-efficient fabrication of intricate nanopatterns using top-down approaches remains a significant challenge. To overcome this limitation, advancements are required across various domains: patterning techniques, real-time and post-process metrology, data analysis, and, crucially, process control. We review recent progress in continuous, top-down nanomanufacturing, with a particular focus on data-driven process control strategies. We explore existing Machine Learning (ML)-based approaches for implementing key aspects of continuous process control, encompassing high-speed metrology balancing speed and resolution, modeling relationships between process parameters and yield, multimodal data fusion for comprehensive process monitoring, and control law development for real-time process adjustments. To assess the applicability of established control strategies in continuous settings, we compare roll-to-roll (R2R) manufacturing, a paradigmatic continuous multistage process, with the well-established batch-based semiconductor manufacturing. Finally, we outline promising future research directions for achieving high-quality, cost-effective, top-down nanomanufacturing and particularly R2R nanomanufacturing at scale.

Keywords: nanomanufacturing; roll-to-roll processing; process control; machine learning; inspection and quality control; metrology

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1 Introduction

Nanotechnology, the manipulation of matter at the atomic and molecular level (1–100 nm), has emerged as a transformative field with the potential to revolutionize numerous scientific and technological domains. A crucial aspect of realizing this potential lies in nanomanufacturing, the scaled-up, cost-effective, accurately controlled fabrication of structures, devices, and systems at the nanoscale (Alexander Liddle and Gallatin 2016). This burgeoning field presents a unique set of challenges and opportunities compared to conventional manufacturing techniques. The rapid, continuous processing offered by high-throughput roll-to-roll (R2R) nanomanufacturing is acknowledged as a critical technology for the development and production of various next-generation devices and flexible electronics (Palavesam et al. 2018; Phung et al. 2021; Zou et al. 2018). Scaling involves overcoming hurdles in areas such as achieving high production rates, ensuring consistent product quality across runs, maintaining precise control over the manufacturing process, optimizing efficiency, while keeping costs competitive.

Analysis (real-time or otherwise) of information across all the relevant time and length scales for high-throughput nanomanufacturing is challenging because of the generation of massive datasets with intricate relationships which exceed the human ability to manually interpret and extract meaningful insights. Machine learning (ML) encompasses algorithmic approaches designed to uncover patterns and relationships within data. In nanotechnology and by extension nanomanufacturing, such techniques find applications in the analysis of large datasets, materials design and discovery, and the optimization of production processes (Brown et al. 2020). Data-driven approaches also support model-based control since building a unified process model based on first-principles approaches for such complex, multi-stage processes is neither straightforward nor practical (Ulbrich and Bloemen Waanders 2018). Furthermore, these processes are likely to exhibit non-stationarity, caused by factors like process drift. As a result, the corresponding process models become increasingly complex and challenging for human operators to maintain and update.

Methods for creating products with nanoscale features fall into two broad categories: bottom-up and top-down. Bottom-up approaches are suitable for manufacturing nanostructures with complex geometries and rely on spontaneous self-assembly processes at the atomic scale. They are driven predominantly by thermodynamic factors. Examples of bottom-up approaches include colloidal self-assembly and DNA-based self-assembly. Self-assembly of colloidal particles can generate superstructures of various dimensions, phases and symmetries and enables the manufacturing of smart materials and devices with highly tunable properties (Z. Li et al. 2022). In DNA-based self-assembly, DNA strands act as template materials which organize disparate nanostructures to construct relatively complex features. While they can produce macroscopic products with nanoscale features, they are inherently slow, and suffer from a lack of long-range order and precise control when operated under non-equilibrium conditions. Bottom-up approaches are promising for laboratory-scale nanofabrication, but improving the yield of self-assembled structures remains a grand challenge. Most bottom-up strategies require external inputs like guide structures, and they often suffer from a relatively high rate of defects that cannot easily be corrected (Fourkas et al. 2021).

Top-down techniques, on the other hand, offer the ability to precisely control the yield and geometry of nanostructures, while ensuring long-range order and high speed. They are therefore better suited for large-scale nanomanufacturing of nanostructures. Integrated circuit (IC) manufacturing represents the salient example of top-down nanomanufacturing. Modern photolithography tools such as extreme ultraviolet (EUV) lithography offer superior resolution and feature quality, capable of achieving sub-10nm critical dimensions. However, its application is primarily focused on high-volume manufacturing of microelectronic integrated circuits due to the exorbitant cost of EUV systems, exceeding one hundred million euros (Zheng et al. 2021). Nanoimprint lithography (NIL), which is an emerging candidate for high-throughput, high-resolution, low-cost nanomanufacturing, uses a hard mold for embossing on a polymer film, either at high temperatures (Thermal NIL) or in the presence of UV radiation (UV-assisted NIL). Nanostructures fabricated by the above-mentioned techniques typically have low aspect ratios and are of lower complexity than in the case of bottom-up methods. Interference Lithography (IL) is a closely related top-down nanofabrication technique that allows for producing 3D nanostructures of arbitrary shape, but the gains in feature complexity come at the loss of processing speed and increased effort for tool setup (viz. coherent optical system with laser source, high-precision mechanics such as work stages for accurate positioning, and beam splitters like Lloyd's mirrors and diffraction gratings).

As of yet, devising a technique or combination of techniques for producing top-down, large-scale, complex nanopatterns at high speed, high quality and reasonable cost remains an open and important scientific and technological question. Addressing this challenge entails advances in feature creation technology, online and offline metrology, data processing and process monitoring and control. Large-scale manufacturing of nanoscale devices comes with many possible applications in electronics, optics, plasmonics, etc. There are many applications requiring large-scale manufacturing of homogeneous nanostructures. Two such examples are the generation of semiconductor nanowires with carefully controlled morphologies for large-scale production of solar cells (Wallentin et al. 2013), and the production of ultrafiltration membranes for water purification. The latter could serve as an ideal candidate for sheet-based nanomanufacturing.

This review explores recent progress in continuous, top-down nanomanufacturing with emphasis on data-driven approaches in the context of process control, while defining the current state-of-the-art in key areas and applications.

Process control in continuous nanomanufacturing involves the following aspects – discussed in greater detail in the subsequent sections (Figure 1):
- Identification of appropriate devices and methods to measure relevant process variables at high speeds while accounting for speed-resolution tradeoff, especially in imaging.
- Modeling of variables representing product quality (which could include feature dimensions, uniformity, etc.) in terms of process and equipment parameters.
- Fusion and interpretation of multimodal data corresponding to multiple length and time scales, to detect equipment (tool) and product defects, if present.
- Inferring process states to determine appropriate control actions based on a developed control law.

We perform a comparative analysis between R2R manufacturing (representative of continuous multistage manufacturing processes) and the industrially well-established semiconductor manufacturing (representative of batch processing), to identify the suitability of control strategies commonly used in the latter for R2R manufacturing. The remainder of this article is organized as follows: Section 2 provides a brief discussion of the history and evolution of

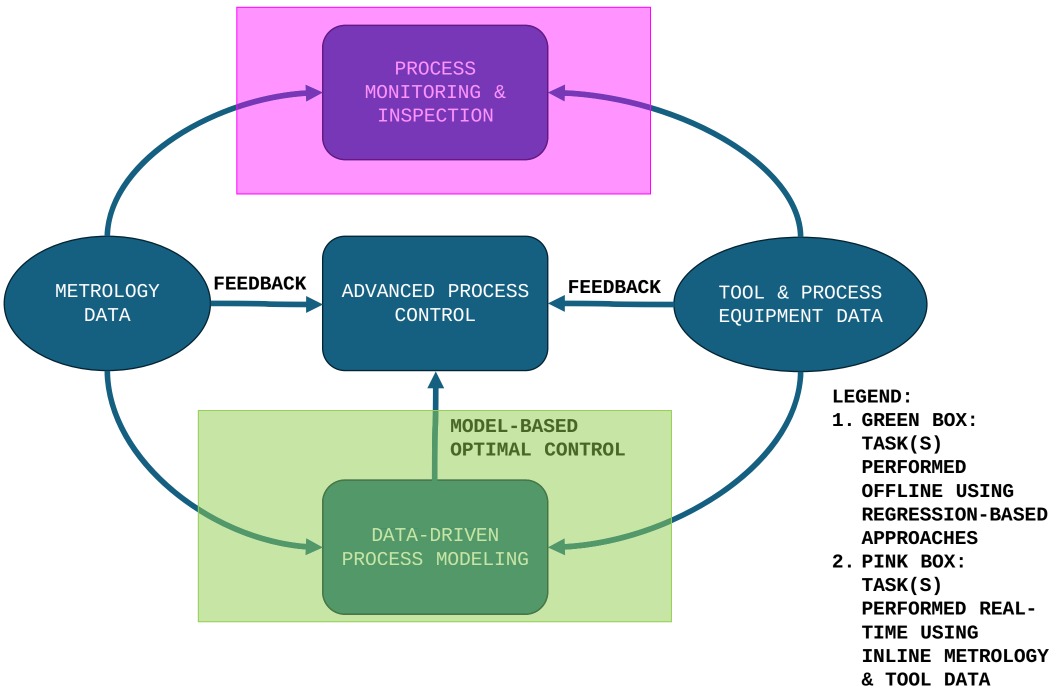

Figure 1: Components of ML-driven process control in a continuous nanomanufacturing framework.

micro/nanofabrication and potential nanopatterning approaches to creating nanoscale features at large scales. Sections 3 and 4, respectively, present discussions of the metrology requirements and overall process control along with process fault detection for the problem of R2R continuous nanomanufacturing. We highlight the use of data-driven approaches to solve relevant problems in R2R manufacturing and semiconductor manufacturing as reported in the literature. Section 5 provides concluding remarks along with suggestions for future research directions.

2 Evolution of micro/nanofabrication techniques, transition to continuous nanomanufacturing

The field of micro/nanofabrication has its roots in the 1950s–1960s, heavily influenced by the needs of the semiconductor industry (Campbell 2001; Jaeger 2002; Plummer et al. 2000). Early techniques focused on photolithography and thin film deposition for microfabrication. The miniaturization of transistors, a key driver of modern electronics, has relied heavily on advancements in photolithography. Over the past several decades, this technique has become the cornerstone of IC fabrication. Photolithography involves transferring a pattern from a photomask onto a substrate, usually a silicon wafer, using light. The process starts with the application of a photosensitive material called photoresist on the substrate. When exposed to light, the photoresist undergoes a chemical change, which allows the subsequent selective removal, via development, of either the exposed or unexposed regions, depending on the type of photoresist used, to create the desired pattern on the substrate. The resolution of the patterns created by photolithography is determined by the wavelength of light used, with shorter wavelengths allowing for finer features. The process can be repeated multiple times to build complex structures, such as those found in ICs. Advancements in lithography, such as deep ultraviolet (DUV) photolithography, enabled the creation of smaller features and more complex integrated circuits. The 1990s brought forth soft lithography and nanoimprint lithography (NIL) (Chou et al. 1996), perhaps being the first step towards continuous patterning of 2D or low aspect ratio 3D nanostructures. Additionally, there is an emphasis on developing scalable nanomanufacturing methods, 3D micro/nanofabrication techniques, and novel materials with unique properties at the micro/nano scale.

The unique properties of periodic 3D nanostructures, stemming from their micro/nanoscale features, have long been a subject of research. Photonic crystals, for instance, exhibit tailored dispersion behavior due to their periodic dielectric profiles. This photonic bandgap allows precise control over light transmittance and reflectance by manipulating the structural periodicity (Campbell et al. 2000; Krauss et al. 1996; Lin et al. 1998; Noda et al. 2000; Qi et al. 2004). Additionally, the high surface area-to-volume ratio of these nanostructures makes them ideal for applications in fast-charging battery electrodes and solar cells (Fan et al. 2009; Zhang et al. 2011). Beyond photonics and energy applications, 3D nanostructures offer intriguing mechanical advantages. They circumvent limitations inherent to macroscale materials, enabling the design of mechanical metamaterials with groundbreaking properties. Studies have demonstrated that periodic nanoarchitectures, or nanolattices, display superior recoverability (Bagal et al. 2017; Jang et al. 2013; Meza et al. n.d.) and unconventional behaviors like negative Poisson's ratio or stiffness (Evans 1991; Lakes 1987; Lakes et al. 2001). Furthermore, they exhibit superior scaling of stiffness and strength with reduced density compared to random porous microstructures (Lee et al. 2010). Nanolattices can also exhibit interesting properties in other physical domains, including refractive indices close to unity (Zhang et al. 2015), improved light trapping (Zhang et al. 2017), and exceptionally low thermal conductivity (Dou et al. 2018). Widespread societal benefits from such impactful advances hinge on breakthroughs in large-scale nanomanufacturing.

Existing top-down approaches to fabricating 3D nanostructures include Focused Ion Beam (FIB), two-photon polymerization (TPP) (Cumpston et al. 1999), and electron-beam lithography (EBL) (Vieu et al. 2000). These methods achieve high-resolution patterning through a layer-by-layer writing approach. However, a drawback common to these techniques is their serial nature, requiring point-by-point scanning, which significantly limits their throughput. In contrast, near-field holographic lithography processes (Kagias et al. 2023; Nesse et al. 2019; Paik et al. 2020) can produce 3D nanoscale features of desired shapes in a single light exposure using sub-diffraction metasurface photomasks. Therefore, higher throughputs can be expected upon their integration with R2R setups. The complexity of the 3D structures can be further enhanced by allowing for the registration of 2nd and higher order diffraction patterns on photoresists (Chang et al. 2011; Zhang et al. 2013).

The most economical large-scale processes are continuous (note that IC production is largely a batch process) in nature and consequently, it is of interest to develop a continuous nanomanufacturing process. The focus of this review is on Roll-to-Roll (R2R) technology owing to its simple transport principle and continuous nature of the manufacturing process (Figure 2). However, we note that there are several transport principles in the substrate (web)-based manufacturing paradigm, such as sheet-to-sheet, sheets-on-shuttle and roll-to-sheet (Willmann et al. 2014). Within this paradigm, a planar (2D) substrate undergoes a series of processing steps that result in the creation of nanoscale features at its surface. This creates a system with two crucial length scales: the feature size at the nanometer scale and the substrate dimension at the centimeter scale. These fundamental characteristics give rise to specific challenges in feature creation, process monitoring and control.

For example, Kagias et al. (2023) observed depth-dependent variation of periodic 3D nanostructures fabricated using IL (with diameters around 460 nm, lateral periodicity of 900 nm on a 30 µm-thick sheet) due to thermal and chemical gradients in the photoresist material during post-exposure development. A key process control challenge in this process is to minimize the occurrence of such variations which also impact material properties of the nanostructures. Certain parameters would need to be more tightly controlled than others, which is usually dictated by

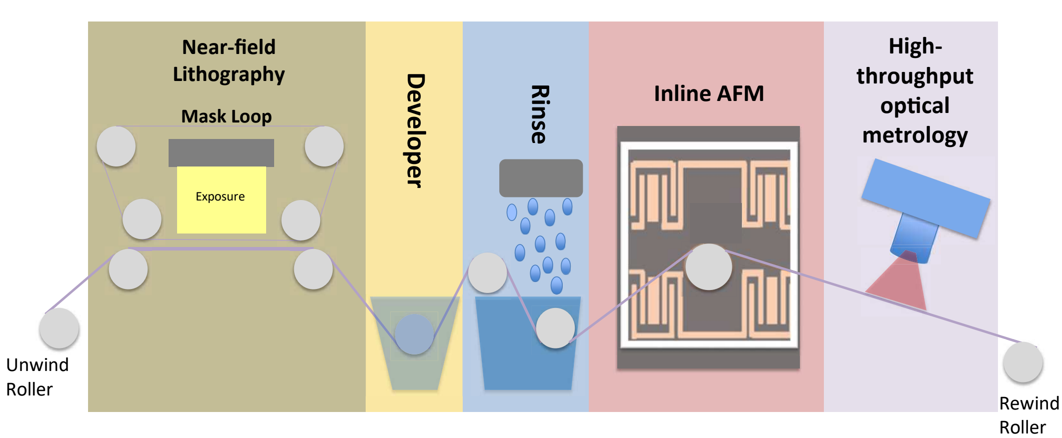

Figure 2: Schematic of a possible framework for R2R nanomanufacturing using near-field holographic lithography. One rewind roller, one unwind roller and several idler rollers are shown. Tension in the conveyed web is controlled by the winder but is typically adjusted by one or more tension rollers located within the R2R machine.

the final product utility. For example, the functionality of printed electronics crucially depends on high layer homogeneity (Su et al. 2019), while multi-layer devices with thin dielectric materials require tight control of surface roughness (Song et al. 2022). Manufacturing ultrafilters demands a narrow pore size distribution while the exact dimensions of pores may not be of much relevance.

3 Metrology

Metrology, as a constituent of nanomanufacturing, allows for inspection of created features and amounts to devising and implementing sensors with nanoscale precision. R2R processing can drastically increase throughputs and reduce overall production costs, but these effects can be nullified if one waits until after a roll of finished product is made before taking measurements on the produced nanostructures. In the context of nanomanufacturing, advances in imaging and image analysis form a cornerstone of automated process control. The nature of processes and products in nanomanufacturing makes quality measurement a nontrivial task. Since effective quality control must be done at rates commensurate with web speeds in R2R manufacturing, metrology must work close to real-time. Moreover, such inline techniques must also be nondestructive, and fairly insensitive to the rapid motion of the object in focus and vibrations in the substrate (Maize et al. 2023).

3.1 Machine learning and metrology in R2R manufacturing

Measurement usually involves a speed-accuracy tradeoff, which becomes highly relevant in the case of metrology in R2R manufacturing. For a given substrate speed, there also exists a tradeoff between the width of the web that can be monitored and measurement resolution. With increasing web width, capturing the entire width of the web in a single measurement is expected to become challenging (Maize et al. 2023).

Techniques such as line scan imaging, hyperspectral imaging and laser scanning which are well established in R2R processing also show promise for use in continuous nanomanufacturing. In imaging systems, particularly spectral cameras, there is a trade-off between spatial and spectral resolution due to limited resources such as detector pixels and exposure time, where enhancing one often compromises the other. Higher spectral resolution significantly increases data volume, impacting processing speed and storage capacity. Efficient data handling techniques are required to balance this with high spatial resolution. Techniques like principal component analysis (PCA) or autoencoder-based compression and parallel processing are essential for efficient data management. Deep learning-based image super-resolution techniques have emerged as a solution to this problem, leveraging high-resolution panchromatic (PAN) and low-resolution hyperspectral (HSI) images to generate high-resolution HSI data (Wang et al. 2021). More specifically, efforts have been undertaken to minimize spectral-spatial distortions in generated HSIs by employing variants of generative neural networks such as the latent encoder-coupled generative adversarial network (LE-GAN) (Shi et al. 2022). Narrowing down spectral data to the relevant range of wavelengths (which is application-specific) can also reduce the sensing cost-resolution trade-off. There is also a growing interest in optical techniques conventionally used in *in-situ* characterization such as angular scatterometry, phase-shifting interferometry, and Raman spectroscopy for inline metrology. The most common parameters measured by or inferred from such techniques include defect density, defect size, film morphology (thickness, roughness, topography), and optical properties (refractive index, reflectance, transmittance). In the case of nanomanufacturing, such techniques can provide insights into consistency and uniformity of fabricated structures, and detect and diagnose defects, although none of them can measure feature sizes at nanoscale resolution. Table 1 provides a list of potential candidates for inline metrology in R2R setups.

There is a need for increased metrology information at higher resolutions to ensure tight quality control at the nanoscale. Atomic force microscopy (AFM), which provides insights into surface topography, is non-destructive, requires hardly any sample preparation and has resolutions on the order of fractions of a nanometer. Connolly et al. (2019) developed an inline single chip-AFM (sc-AFM) framework for a R2R process. It is modular and allows for scaling the number of probes and approach mechanisms to increase the overall throughput at no extra computational cost and a negligible increase in the physical space occupied. However, since the entire web cannot be scanned using this method, the optimal frequency of scanning and locations to be scanned must be determined in order to obtain maximum information about the trends in dimensions of fabricated features across the entire web, while ensuring high production rates. There has been a general interest in the automation of microscopy (Kalinin et al. 2021) in the context of selecting the appropriate imaging focus regions and microscope hardware tuning, which largely remain dependent on human expertise.

Table 1: Optical metrology techniques for R2R nanomanufacturing: potential candidates.

Technique	Measured parameters	Demonstrated (inline) speed	Demonstrated resolution	ML contributions and other remarks
Angular scatterometry (Faria-Briceno et al. 2019)	Reflectance as a function of angle of incidence	10 cm/s	10 nm (can only be achieved in highly periodic structures, a specialized case that allows for resolution much better than the diffraction limit)	Reflectivity-incident angle plots can be used to monitor feature trends in the machine direction for *periodic structures*. ML approaches can help select informative wavelengths for analysis, reducing reference library search times (Sabbagh et al. 2023b)
Inline phase-shifting interferometry	Thickness, surface topography, displacement	2.5 cm/s	Lateral: 1 μm for field of view of 4 mm Vertical: 5 nm	Insensitive to vibrations in web and enables single-shot measurements by extracting phase information from a single interferogram, overcoming limitations of traditional methods that require multiple exposures. Zhang et al. 2021 developed a one-to-multiple deep learning framework to generate the equivalent of multiple phase-shifted interferograms from a single inline interferogram
Hyperspectral linescan imaging	Composite image over a wide range of wavelengths (infrared/visible/UV), representing the entire web or parts thereof	Not demonstrated yet	1.2 μm	Data driven approaches (such as k-nearest neighbors and PCA-based schemes) can be used to determine feature dimensions and select useful wavelengths (Gawlik et al. 2020, 2021; Yue et al. 2000)
Raman spectroscopy	Image part of the web. Principle is similar to angular scatterometry, with higher resolution but much lower web speeds	120 μm in 5 s (10X speeds using wavelet-based approaches (Yue et al. 2018)	1 μm	(Yue et al. 2017, 2018, 2020) developed a single overall metric to track feature trends along the machine direction, with deep learning-enabled Raman spectroscopy improving data acquisition rates (Horgan et al. 2021)

A related case of determining scanning trajectories and locations can be found in paper manufacturing, an industrially well-established problem. The machine direction (MD) and cross direction (CD) profiles of paper properties are measured by a scanning gauge containing an array of sensors (Astrom 1967; Dave et al. 1997; Dumont et al. 1993; He et al. 2015; Rippon et al. 2019; Stewart 2000; Valenzuela et al. 2003). Sensors are typically guided in the CD while the paper moves at high speeds, whereby the sampled points form a diagonal trajectory on the paper sheet. Dedicated MD and CD control systems are employed to address temporal and spatial variations, respectively. Due to the zig-zag sampling trajectory, a mix of MD and CD variations is embedded in the measurements. Hence, their separation becomes an important task to generate separate control inputs, while accounting for possible aliasing effects (Rippon et al. 2019).

3.2 Machine learning and metrology in semiconductor manufacturing

Semiconductor manufacturing is a complex process that requires monitoring of several inter-related critical process parameters from the initial stages of production to the packaging of the final product. It comprises four main stages: wafer fabrication, wafer inspection, assembly, and final testing. In the fabrication stage, wafers undergo numerous (often hundreds) sequential processing steps (deposition, lithography, etching, implantation, polishing, etc.) in batches (groups of tens of individual wafers). The entire manufacturing process may require up to three months to produce a chip. Therefore, it may take *months* since the commencement of operations to determine product yields – requiring *soft sensing* in the intermediate stages to achieve effective process control (Qin et al. 2006; Su et al. 2007).

Table 2: Comparison of dimensional metrology methods in semiconductor manufacturing.

Technique	Measured parameters	Resolution (lateral and vertical)	ML contributions and other remarks
Critical dimension-scanning electron microscopy (CD-SEM)	Image portions of the wafer (wide field of view of 50 nm to 10 mm)	~ 0.3 nm	Yields top-down images providing critical IC dimensional parameters such as linewidth, edge roughness (Mack and Bunday 2018), and contact holes (Bunday et al. 2018); requires that sample surfaces be conductive and be placed in high vacuum. Image super resolution using generative-adversarial networks has been explored aiming to achieve high-resolution images with minimal electron dosage and sample damage (Liu et al. 2019)
Scatterometry	Reflectance as a function of wavelength/angle of incidence	Model-dependent; ~ 1 nm (vertical and lateral): Note the higher resolution compared to that achieved in inline R2R setups	Provides data-driven model-based estimates of overlay effects (den Boef 2016; Peled et al. 2018), critical dimensions and optical constants of periodic patterns. Fast and non-destructive, allows for inline measurement. ML techniques have been widely applied to monitor critical dimension variations and solve the inverse problem of determining dimensions from spectra (Liu et al. 2022; Lucas et al. 2018)
Atomic force microscopy (AFM)	3D surface topographical maps and mechanical properties such as stiffness and adhesion forces	1 nm lateral, 0.1 nm vertical	Little to no sample preparation required, non-destructive, inline compatibility; nanometer-level measurement accuracy. Involves raster-scanning a sharp probe tip across the sample. A recent work explored the use deep learning approaches to autonomously perform AFM instrument initialization, surface imaging, and image analysis (Kang et al. 2023)
Spectroscopic ellipsometry	Ratio of the amplitude of *p-polarized* to *s-polarized* reflected light and the phase difference between the two polarizations	Model-dependent	Used to analyze thin films, measure layer thickness, refractive index, and optical constants, and study surface roughness and composition; mainly used for depth monitoring for 3D structures to ensure precise control over film thickness. Recent works (Li et al. 2021; Liu et al. 2021) have investigated the use of deep learning frameworks to accelerate the analysis of ellipsometry data for high-throughput experimentation

Due to cost and time constraints, wafer metrology is performed on a statistically representative subset of wafers after key processing steps. Metrology focuses on measuring various properties of materials and processes, including thickness, electrical resistance, critical dimensions (key features on a microchip), alignment of layers, particle contamination, and the rate of material removal during etching. *Ex-situ* metrology plays a crucial role at each stage of semiconductor manufacturing as it is the predominant source of product quality information before and after that process. This information is used to determine whether the current processing conditions and tools need to be adjusted in subsequent runs. Within the manufacturing tool itself, conditions like temperature, pressure, flow rate, and electrical current are recorded at much faster timescales (in the order of milliseconds) (Su et al. 2007). Table 2 summarizes and compares key dimensional metrology techniques used in semiconductor manufacturing.

Metrology delay in semiconductor manufacturing processes is inevitable, which can adversely impact process control performance. Another inherent limitation of such metrology approaches to guide decision making is the assumption that a few samples are representative of the whole lot, which can make defect detection very challenging since only a small fraction of wafers is expected to be faulty.

To ensure efficient production, significant efforts are underway to improve the speed and cost-effectiveness of metrology measurements. Researchers have developed techniques to speed up SEM imaging by using optimized beam scanning approaches (Cizmar et al. 2011; Kruit et al. 2016; Sunaoshi et al. 2016). Such methods only acquire the data necessary for the desired image, reducing acquisition time. Additionally, deep learning algorithms are being implemented to significantly improve both the speed and quality of images by removing noise, with recent advancements including patch-based algorithms for denoising low-dose SEM images (Lazar and Fodor 2015) and reconstructing high resolution AFM images from low resolution, high scan speed data (Natinsky et al. 2024). In such applications involving ML for signal denoising, neural networks are typically trained on low-resolution (LR) and high-resolution (HR) data pairs to learn effective representations that reconstruct low-noise outputs from high-noise inputs. Horgan et al. (2021) show that deep learning significantly outperforms traditional spectral smoothing algorithms like Savitzky–Golay (SG), wavelet, and PCA, enabling effective reconstruction of Raman signatures from low SNR spectra. In this context, we make the distinction of the noise in low SNR signals, resulting from high data acquisition speeds (the noise signal of interest), from noise present in the training data (which is universal). Another approach utilizes machine learning with non-linear anisotropic diffusion for denoising images specifically intended for electron tomography applications (Staniewicz and Midgley 2015).

In optical metrology, extracting feature dimensions and thicknesses from measurements falls under the mathematical category of an inverse problem (Xie et al. 2019; Sabbagh et al. 2023a,b). This problem seeks to determine the "causes" from the observed "effects". Inverse problems often lack analytical solutions and machine learning approaches such as deep neural networks and regression-based methods have been used (Barkhordari et al. 2024; Liu et al. 2021; Zhu et al. 2024) to iteratively identify a set of optical parameters that best match the measured data.

Virtual Metrology (VM) in semiconductor manufacturing leverages machine learning and statistical methods to predict crucial properties of wafers without direct physical measurement (Kang and Kang 2017; Kang et al. 2009, 2011). This bypasses the need for expensive and time-consuming traditional metrology tools. These models are typically constructed by training ML algorithms on historical process data, including logistical and process parameters, and corresponding physical metrology results. VM relies on data collected from sensors embedded within the processing equipment itself. These data include temperature, pressure, and power consumption. Example algorithms are multivariate regression and multi-level models with regularization. Once trained, the VM system can estimate desired parameters like critical dimension or layer thickness for each wafer based solely on sensor readings, enabling real-time process monitoring and optimization. Due to the high dimensionality of VM feature spaces, arising from numerous high-frequency process sensors, dimensionality reduction is critical. This preprocessing step can be achieved through feature extraction, with convolutional neural networks (CNNs) dominating this area as of 2020 (Dreyfus et al. 2022).

VM approaches have been successful in overcoming the sparsity challenge, a major hurdle in traditional process monitoring where only a small subset of wafers is measured with physical metrology tools (Maggipinto et al. 2019). developed a deep learning approach specifically for VM in the etching step of wafer fabrication. Their method leverages optical emission spectral (OES) data, which capture the light emitted during the etching process. By feeding these data into a deep neural network, the VM system can infer the etch rate for each wafer within the batch. This information is crucial for quality assessment, as an uneven etch rate can lead to device defects.

Ensuring sustained high performance for a VM system over long durations is difficult due to temporal variations in the underlying data characteristics. These variations can be caused by several factors, including internal process drifts, equipment disturbances, external environmental fluctuations, and routine maintenance interventions. To counteract this performance decline, VM systems must integrate real, physical metrology data into their workflows for practical deployments. However, increasing the dependence on actual metrology measurements incurs a significant cost penalty. Consequently, a critical aspect for successful VM implementation is the development of a strategic approach for selecting wafers that require actual metrology measurements (Baek et al. 2014; Hyun Baek et al. 2014; Kang and Kang 2017; Lu et al. 2014).

3.3 Machine learning enabled-multiscale and multimodal metrology

Understanding interactions among components of complex systems and processes requires sensing many types of process variables (multiple modalities). Such measurements oftentimes span multiple length and time scales, as in the problem of R2R nanomanufacturing, and data fusion is concerned with meaningfully combining and extracting such multimodal data which typically complement one another (Figure 3). Recent years have witnessed an unprecedented surge in the development of deep learning technologies to perform multimodal data fusion to study complex systems, taking advantage of the complementarity (providing insights which are unobservable if data from a single sensor are analyzed) and redundancy (particularly useful while dealing with noisy or missing data) of heterogeneous process data (Gao et al. 2020; Zhao et al. 2024).

For example, Li et al. (2022) performed *in-situ* product quality monitoring in a metal 3D printing process using heterogeneous sensor measurements. A convolutional neural network (CNN)-based data fusion approach was adopted to combine the disparate yet relevant signals from a digital camera, a microphone, and a photodiode, measured at different rates. The main bottleneck in these applications is the scarcity of training data. This limitation arises because acquiring sufficient data necessitates running the actual process for extended periods, which can be time-consuming and resource intensive.

Lu and Jayaraman (2023) demonstrated the use of paired variational autoencoders (Pair-VAE) for correlating low-resolution small-angle X-ray scattering (SAXS) data with high-resolution scanning electron microscope (SEM) images, where both originated from the same sample location. This approach holds promise for implementing inline metrology in nanomanufacturing. However, a key challenge arises when attempting to integrate inline optical metrology data, which inherently represents larger sample areas compared to high-resolution microscopy images. To ensure signals correspond to the same process region, techniques like fiducial markers (Potočnik et al. 2022) are valuable for automated object tracking in R2R manufacturing. However, these markers may not be suitable for high-resolution AFM due to inherent size limitations.

Sensor data fed into fusion systems can be corrupted by various failures, arising from process or equipment faults, leading to inaccurate estimates if fused with valid data (Kumar et al. 2006). Sensor redundancy, where multiple sensors monitor the same phenomenon, allows for cross-checking and filtering out faulty data points. Furthermore, robust fusion algorithms can be implemented that are less sensitive to outliers and can down-weigh the influence of potentially corrupted data in the final estimate. The significant disparity in data acquisition rates between high-resolution imaging and faster optical metrology necessitates careful data fusion techniques to prevent out-of-sequence integration. Out-of-sequence measurements (OOSM) can lead to inaccurate controller inputs due to mismatched information, potentially causing fabrication errors and unreliable device performance. These OOSM can be misinterpreted as

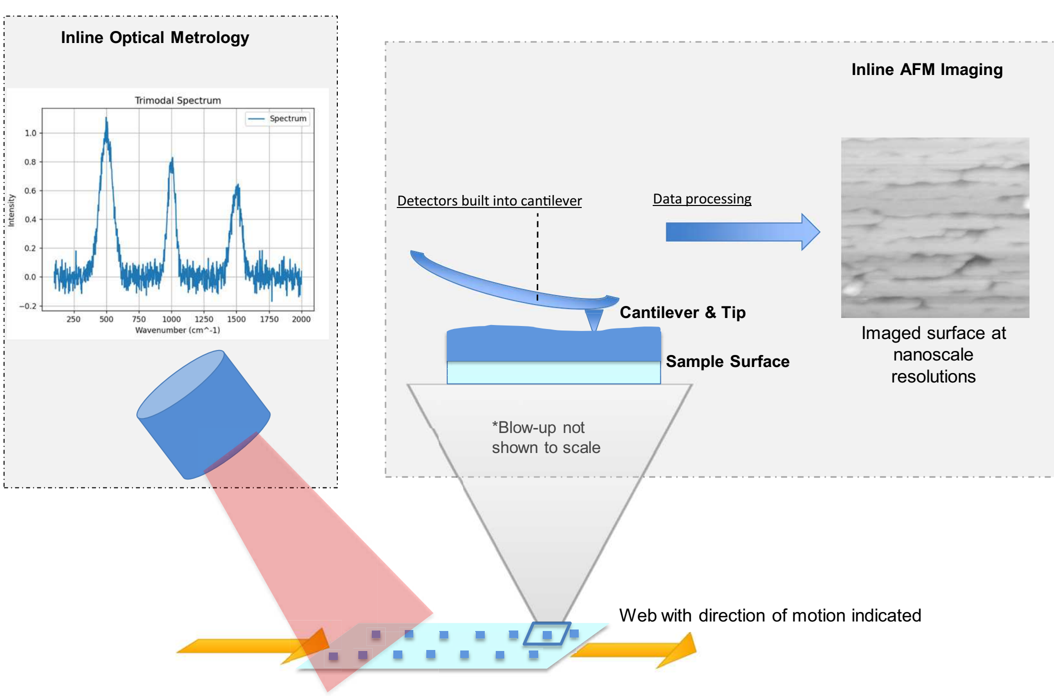

Figure 3: A schematic illustrating the multiscale, multiresolution nature of the metrology framework. Optical metrology techniques operate at high speeds, yielding web-wide information at lower resolutions. The peaks in the intensity plot correspond to wavelengths that are strongly reflected by the material. Inline AFM provides local (marked by blue squares, see blow-up) nanoscale feature-level information at lower speeds.

inconsistencies by the fusion algorithm. The challenge lies in effectively incorporating these often-older data to update the current estimate while accounting for the correlated process noise between the current time and the time of the delayed measurement (Khaleghi et al. 2013). A simple solution is discarding OOSM, but this leads to information loss and performance degradation if OOSM is prevalent (Kim et al. 2018).

Furthermore, the data fusion process can generate "out-of-distribution" (OOD) features when combining sensor signals with inherent inconsistencies. This is particularly relevant for neural networks, which have been shown to assign high confidence to unexpected OOD inputs that fall outside the data they were trained on (Ming et al. 2022). This highlights the critical role of OOD detection in data fusion with neural networks. By identifying and handling these unknown OOD inputs, the algorithm can apply safety precautions and avoid making unreliable predictions based on out-of-scope data. Further research on synchronization, data validation, and advanced fusion methods are crucial for ensuring reliable control based on a comprehensive picture of the nanoscale features.

3.4 Key findings for R2R nanomanufacturing: metrology

- An inherent limitation of sensors with nanoscale resolution is that they can only cover a limited (from a macroscopic perspective) area of the substrate, and additional effort is required to obtain relevant nanoscale information for the entire substrate at all times (i.e., inline). One possible solution to this problem is using an inline AFM for acquiring real-time nanoscale data directly during the manufacturing process since it requires minimal sample processing and causes minimal damage.
- While electron microscopy offers unparalleled resolution for analyzing nanostructures, it presents significant drawbacks in the context of R2R manufacturing with flexible substrates. The technique's limitations in sample size restrict its applicability for monitoring large, continuous webs. Additionally, the extensive preprocessing steps required to prepare samples for electron microscopy and requirements on vacuum during measurement, are incompatible with the real-time, in-line process control needed in R2R setups.
- Increased dependence on inline and virtual metrology: It is infeasible to isolate samples of fabricated nanostructures for offline inspection until the final products are produced. This contrasts with semiconductor manufacturing where actual metrology data from intermediate process steps are more easily available. Consequently, offline metrology data could lag significantly behind upstream fabrication processes. This leads to increased dependence of VM models on inline metrology data. Hence, it is essential to obtain feature dimensions (inline) with high accuracies at nanoscale.
- The use of machine learning approaches to enable improved inline process monitoring can take the following forms:
  - Learning and filtering environmental factors – enabling image denoising to reconstruct high quality images while improving data acquisition speeds.
  - Integration of multiscale and multimodal data for process monitoring, which may not be feasible using first principles modeling approaches.
  - Fast computation of solutions to inverse problems using optical metrology information.
  - Virtual Metrology techniques can be adopted due to the sparse nature of high-resolution *in-situ* and *ex-situ* measurements (actual metrology data) in a continuous R2R setup. Moving window approaches can be explored to enable automated model training for inline implementation, to counteract the effects of process non-stationarities/drifts.

4 Machine learning for process control

Limitations in the mechanistic understanding of the complex interactions among materials, pressures, temperatures, and chemicals involved in nanofabrication become a critical hurdle to achieving tight control, especially as device features shrink and performance demands increase. Atomic layer processes are typically nonlinear, and several methods have been investigated to integrate nonlinear input–output relationships into run-to-run (RtR) control models in the semiconductor industry (Smith and Boning 1997). Atomic layer deposition (ALD) exemplifies this, using sequential, self-limiting surface reactions with reactive gaseous precursors and purge cycles to control film thickness and composition precisely. The process's nonlinearity stems from the interactions among surface reactions, precursor adsorption/desorption, and transport phenomena. Nwanna et al. (2022) identified a nonlinear relationship between flow rates and film deposition rates, with lower purge flow rates enhancing mass fraction distribution and deposition rates due to increased precursor residence time. Additionally, the rate of surface reactions is nonlinearly dependent on temperature, another potential manipulated variable (along with flow rates) for process control. An approach to nonlinear process control, as proposed by Yun et al. (2022), involves linearizing a pre-generated, non-linear dataset using a sigmoidal-like, median-effect equation. This essentially transforms the complex data into a format that a linear R2R control model can work with. Another study investigated and compared two different run-to-run (RtR) control strategies for spatial thermal atomic layer etching of aluminum oxide thin films. One approach utilized a control law based on an artificial neural network (ANN) in feedback with the actual process to achieve disturbance rejection capabilities, while the other relied on a traditional Exponentially Weighted Moving Average (EWMA) method (Tom et al. 2022). It was found that while a lower EWMA weight effectively minimized noise and variance during shift disturbances, it failed to achieve the desired target when subjected to a non-deterministic drift disturbance in simulations. In contrast, the ANN-based RtR controller successfully mitigated the effects of both mild and severe shift disturbances, as well as drift disturbances.

A key difference between nanomanufacturing process control and traditional chemical process control lies in the higher frequency of tool maintenance and repair. This is driven by the increased sensitivity of nanomanufacturing processes to environmental contaminants. Even minor imperfections on tools or the presence of foreign particles can significantly impact the precise control required at the nanoscale. Product specifications should be regulated using feedback control to compensate for tool degradation.

In the context of manufacturing, system identification plays a vital role in making production scheduling decisions, enhancing control policies, and adapting to changes in process requirements and system structures. By utilizing approaches like machine learning algorithms or genetic programming, system identification can address the shortcomings of traditional approaches and offer more accurate models that reflect the dynamic nature of manufacturing systems (Denno et al. 2018). There is increasing interest in approaches that decrease reliance on process models for controlling complex dynamical systems. Traditional modeling techniques can fail to capture the multiscale nature of systems, especially in scenarios involving large state and action spaces and multiple inputs and outputs. The use of simplified linearized process models often becomes necessary, either due to the impracticality of constructing nonlinear models or to minimize the issues of computational burden and solution intractability associated with nonlinear models (Spielberg et al. 2019).

Developing accurate process models in industrial settings is particularly time-consuming and challenging, and it frequently requires model reidentification runs that can halt plant operations for extended periods. Reinforcement learning (RL) offers substantial advantages in addressing these challenges through its model-free approach, which learns action policies (i.e. an implicit control law) directly from interactions with the environment, thereby obviating the need for precise pre-existing models. RL's adaptability enables it to manage environmental or process dynamics changes without requiring explicit model updates, making it well-suited for handling multiscale disturbances and complex dynamics. Advanced RL algorithms can effectively manage continuous state and action spaces, facilitating more precise and nuanced control strategies (Cai and Vasile 2021).

Deep RL (DRL) techniques offer inherent flexibility and generalization capabilities, reducing the need for constant parameter tuning and control law redesign (Bougie et al. 2022). Additionally, RL can be integrated with other methodologies, such as wavelet analysis, to bolster its capacity to manage multiscale disturbances and complex dynamics (Ganesan et al. 2007). In contrast to model-based approaches such as robust MPC which incorporate model uncertainties directly into the optimization framework, DRL controllers continuously update network parameters at each sampling instant based on their latest observations, thus not requiring external tuning efforts (Spielberg et al. 2019).

On the other hand, the assumptions of discrete-time dynamics and the availability of complete state information in RL-based controllers hamper their implementation in the industry. In the context of nanofabrication, Leinen et al. (2020) demonstrated the use of RL to autonomously manipulate single molecules despite challenges posed by environment non-stationarity and limited state observability at the nanoscale. Implementing RL-based controllers in continuous nanomanufacturing requires careful selection of variables for the RL state vector. This selection is critical as it influences the number of trials needed for training and the feasibility of achieving effective control. If the chosen state description does not adhere to the Markovian property, successful control may not be guaranteed. At the nanoscale, the use of hybrid learning approaches is recommended (Leinen et al. 2020), by integrating fundamental process physics knowledge in simulations to steer the agent's exploration of potential solutions.

4.1 Machine learning for process fault detection and classification

Typical inline metrology datasets include images and spectra. ML algorithms can help identify empirical correlations within such data at fast rates once models are trained off-line, prior to their deployment. The use of supervised learning approaches such as convolutional neural networks for the analysis of images (which are inherently spatially correlated) has been successful (Guo et al. 2017; Hussain et al. 2019; Li et al. 2014).

Deep learning has been widely employed for image recognition, enabling automatic image categorization and labeling. For example, Anand et al. (2021) focused on leveraging deep learning techniques to enhance the inspection process and reverse defects in nanopillar arrays, crucial for advancing next-generation transistors. By optimizing the use of a small dataset of defective nanostructures, this method demonstrated improved efficiency in identifying and rectifying collapsed nanostructures, offering a versatile platform for high-throughput inspection and defect elimination in 3D nanofabrication processes. In a production setting, it is highly likely (and preferable) that the number of defective samples will be small. Another example involves the use of predictor-corrector CNN models to address the challenge of high sensitivity of photonic devices to nanofabrication process variations (Gostimirovic et al. 2023). By automatically correcting design layouts before fabrication, these models aim to enhance the fabrication fidelity of photonic devices, ensuring higher quality and performance in the final products. Prior to deploying such models on running processes, they must be trained offline on large, high-quality datasets which can take a considerable amount of time. A key challenge lies in accounting for potential class imbalance in training datasets while building neural network-based classifiers for defect classification, especially since the amount of “normal” operating data would far exceed the amount of “faulty” process data.

Advances in process design alone may not guarantee product quality due to the complexity and nature of uncertainties and disturbances involved in multistage manufacturing processes. Fault detection and classification (FDC) in manufacturing processes is therefore crucial for ensuring products with high standards and the reliable and safe operation of industrial equipment. Since unexpected deviations in sensor measurements from actual values can degrade control performance, determining if a fault has occurred using additional data (besides metrology) is important. This means that information from the operation of the machines (speed, tension, temperature, power, etc.) and controllers can be fused with metrology data to make the determination (typically binary) regarding whether the product is “normal” or a fault has occurred – the construction of a decision rule, for which statistical methods such as multivariate control charts are among the most widely used.

A conventional approach to FDC involves an assumption that the observations fall within a multidimensional Gaussian distribution with unknown mean and variance structure. In such a scenario, the Hotelling t^2 statistic is the most appropriate method to detect faults arising from shifts in means of observations. Prior to the implementation of such a rule, it is recommended to reduce the dimensionality of data using projection methods such as principal component analysis (PCA), which finds place in a host of literature concerning FDC in a wide range of industrial processes (Russell et al. 2000; Venkatasubramanian et al. 2003). The next logical step in FDC involves the determination of the cause of the detected fault. A widely used solution is the contribution plot, which shows the contribution of each process variable to the statistic calculated. A high contribution of a process variable usually indicates a problem with this specific variable (Westerhuis et al. 2000). Smith et al. (2024) present a Riemannian framework for material shape analysis. This domain-agnostic, computationally efficient method integrates well with common data analysis techniques (statistical moments, dimensionality reduction, statistical process control). Unlike ML-based image analysis, it does not require large amounts of well-sampled training data particularly containing substantial defective samples.

4.2 Process control in R2R manufacturing

Continuous manufacturing in R2R setups on flexible substrates calls for increased automation in controlling the size, uniformity, and quality of nanostructures. Implementation of real-time process control in such a setup needs to overcome challenges such as limited physics-based process understanding and the unavailability of *in-situ* measurements of important process variables. The continuous nature of R2R processes means that variations at one stage can propagate downstream, potentially leading to cumulative quality deviations. Identifying the true source(s) of such variations using metrology information predominantly acquired downstream in the processing line is critical to ensuring tight product quality control.

Graff and Djurdjanovic (2022) investigated the use of single input-single output (SISO) control strategies on an R2R-based physical vapor deposition process, to control the deposited film thickness with sputtering power as the input variable. Spectroscopic measurements on finished portions of the web aided in film thickness estimation and the modeling of film thickness as a function of input power. This highlights the need for reliable data-driven process models to achieve control of desired variables for all such constituent processes in R2R nanomanufacturing. In a more realistic scenario involving multiple manipulated and controlled variables (Multiple input-multiple output; MIMO), there typically exist non-linear and interactive relations among the variables, and consequently, the complexity of model building increases. Since measurements of outputs are expected to be made downstream of the actual process, they can lag the process significantly.

Just like metrology, control in continuous nanomanufacturing must also consider multiple and disparate time and length scales. This requires tight control of web tension and web speed at the macroscopic level and feature dimensions at the nanoscale. The reader is referred to existing literature on of web tension, speed, and position control problems (Feng et al. 2021; Lee et al. 2020; Raul and Pagilla 2015; Yan and Du 2020). High radial stresses induced during web take-up are recognized to be a significant contributor to post-winding product deformation. Conversely, insufficient radial stresses can lead to web loosening. As winding tension is the sole controllable parameter influencing the radial stress state within the web, these considerations establish critical constraints on the web tension control strategy. The maximum operating web speed should enable the slowest step of the process to be operated robustly in the case of a single-pass, single-line operation. Otherwise, the operation can be split into more than one line, each operating at a different speed (multi-line), or by running the web multiple times through the same line at different speeds, performing different operations in each pass (multi-pass). However, a multi-pass/multi-line operation incurs additional cost and lowers throughput, and hence should be avoided to the extent possible. Additionally, a multi-pass operation suffers from another important limitation-that of producing sheets possessing nanostructures with noticeable variations along the CD.

The creation of nanoscale objects is irreversible in that reworks on existing nanostructures are nearly impossible. Therefore, the feedback loop can only ensure improvements to the quality of nanostructures fabricated subsequently (Willmann et al. 2014). Since the large-scale R2R production of nanostructures will be advantageous from a cost perspective only if scrap rates are kept low, an important goal is to minimize the amount of substandard products generated. Much variation in the process arises during start-up and shutdown, and tightening control in these regimes can increase product yields.

Rollers and substrates are critical components of an R2R manufacturing system at the macroscale. In paper production, fluctuations in web tension have been found to impact the rheology of ink deposited on the substrate. Since the post-exposure development of coated polymer films requires the use of a solvent, ensuring its uniform distribution becomes key to the generation of uniform nanostructures, which becomes yet another reason to tightly control web tension. Lateral motion errors can create wrinkles in a moving web subsequently leading to register errors. It was found that tension-related disturbances accumulating in webs through tension transfer could also lead to register errors (Lee et al. 2022).

Most of the current industrial R2R processes are not fully observable due to the lack of sufficient *in-situ* sensors which are cost-effective. Virtual metrology approaches have been proposed (Jin et al. 2019; Shui et al. 2019) which aim to predict spatial variations in substrate characteristics (alignment, speed, tension, etc.) by integrating first principles models based on web kinematics and sparsely available physical sensor measurements. As discussed earlier, increased availability of estimated/measured variables at high spatial and temporal granularities would allow for more reliable detection of faults and/or defects.

The presence of different sampling rates associated with high resolution versus large area metrology techniques lend itself to the use of multirate control strategies (Dai et al. 2022; Li et al. 2020). A single controller regulating multiple outputs sampled asynchronously at different frequencies providing control moves at fast rates, can be designed with the aid of decentralized multirate state estimation schemes (Sun et al. 2024; Zhang et al. 2022). However, a key distinctive feature of the nanomanufacturing problem is that the input variables of such a controller pertain to vastly different length scales, and using a single controller could pose numerical challenges.

At the nanoscale, the metrology techniques reviewed earlier represent the foundation of controlling feature creation and ultimately the quality of the product. The fact that information at the nanoscale is sparse in both space and time poses an inherent challenge to control performance and ensure product quality. Manohar et al. (2018) investigated the problem of optimized sensor placement for signal reconstruction using customized sets of features extracted using data-driven approaches. Here, advantage was taken of the fact that the dynamics of high-dimensional states typically have low-rank representations, which allows for sparse sampling and full signal reconstruction from a small subset of measurements. However, R2R nanomanufacturing would involve sensors with multiple data types corresponding to different scales. A similar challenge arises in the process control of semiconductor manufacturing (described in greater detail below), where actual metrology data from individual processes are available only from measurements made on a few sampled wafers.

4.3 Process control in semiconductor manufacturing

Run-to-run (RtR) control is the most common and established control strategy used at the equipment level using metrology data, where the processing conditions of each equipment are updated after each batch of wafers is processed. RtR fine tunes recipes of either the previous (feedback) or next processing step (feedforward; to compensate for variations caused in the previous process) to minimize the effects of process drift and variability. While RtR control can compensate for process and equipment drifts through metrology feedback, it cannot compensate for metrology drifts and uncertainties (Qin et al. 2006).

Advanced process control (APC) regulates processes fab-wide, by determining set points for the lower-level RtR controllers based on yield analysis using metrology results obtained from finished wafers – in other words, it aims to maximize yield, while meeting required demands. High yields also significantly decrease manufacturing cycle times, facilitating timely product delivery, thus representing one of the most important performance indices of the manufacturing process. Various studies have proposed models to accurately predict yield that can be used in model-based controllers in APC; reviews of yield modeling approaches for semiconductor manufacturing can be found in (Kumar et al. 2006; Milor 2013).

The APC layer broadly comprises the RtR controllers, VM systems and the fault detection system. Khan et al. (2007) designed a fab-wide control framework by utilizing VM generated for RtR control at the wafer-level. However, a challenge associated with the use of VM lies in maintaining high accuracy of prediction models. In practice, machine conditions change frequently and non-periodically, and it thus becomes imperative to update the VM prediction models periodically. Fan and Chang (2013) developed an integrated APC system for wafer fabrication processes which used a recursive moving window based VM module which was updated whenever new metrology data became available.

Yet another aspect that is closely related to process control is the scheduling of manufacturing operations. The varied nature of operations in semiconductor process flows – from the duration of individual operations to the nature of processing wafers within an individual process – leads to the formation of long queues in front of machines and a non-linear flow of products, thus calling for incorporating production schedule optimization (Mönch et al. 2011). Equipment failures induce downtime and introduce variability in processing times. Critical machines, like steppers, often undergo post-repair test runs followed by a re-qualification process. Re-qualification verifies if the machine meets process tolerances for wafer production. If test runs yield non-compliant wafers, they require rework, further extending the processing queue for subsequent machines (Sarin et al. 2011). Since APC is concerned with the operation of machines and process recipes, the scheduling and control problems are mutually dependent (Baldea and Harjunkoski 2014; Baldea et al. 2015; Caspari et al. 2020; Santander et al. 2023; Tsay and Baldea 2020). The time scales associated with such dynamics are comparable to those of process control, and hence integrating the two problems becomes essential. Implementing APC with a sole focus on product quality control without considering existing schedules can interfere with fab-wide production plans.

4.4 Key findings for R2R nanomanufacturing: process control

Unlike in semiconductor manufacturing where the materials undergo processing as batches of multiple wafers, we can expect processing to take place simultaneously in different sections of a single long roll of the substrate in R2R nanomanufacturing. Figure 4 presents a list of variables of interest in the advanced process control for R2R nanomanufacturing. We share some perspectives on process control and potential challenges that are critical to R2R nanomanufacturing which are listed below:

- Defects in the fabricated nanostructures could arise because of faults in equipment such as rollers, sensors, and photomask (systematic) or due to the contamination of equipment by dust particles (random) – it becomes essential to distinguish one type of fault from the other.
- The continuous nature of R2R nanomanufacturing makes the process amenable to the implementation of continuous-time cascade control strategies.
- Intermediate process steps can be nonlinear and non-stationary, calling for data-driven process modeling for model-based optimal control. Alternatively, data-driven model-free control approaches such as dissipativity learning control (DLC) (Tang and Daoutidis 2021) can be explored to directly extract control-relevant information from data. While potentially less comprehensive than a complete dynamic model, such an approach exhibits a more direct relationship with control performance.
- Alongside inline metrology on samples performed after resist development, machine vision systems can be placed to monitor the progression of the photoresist material through intermediate processes up to and including

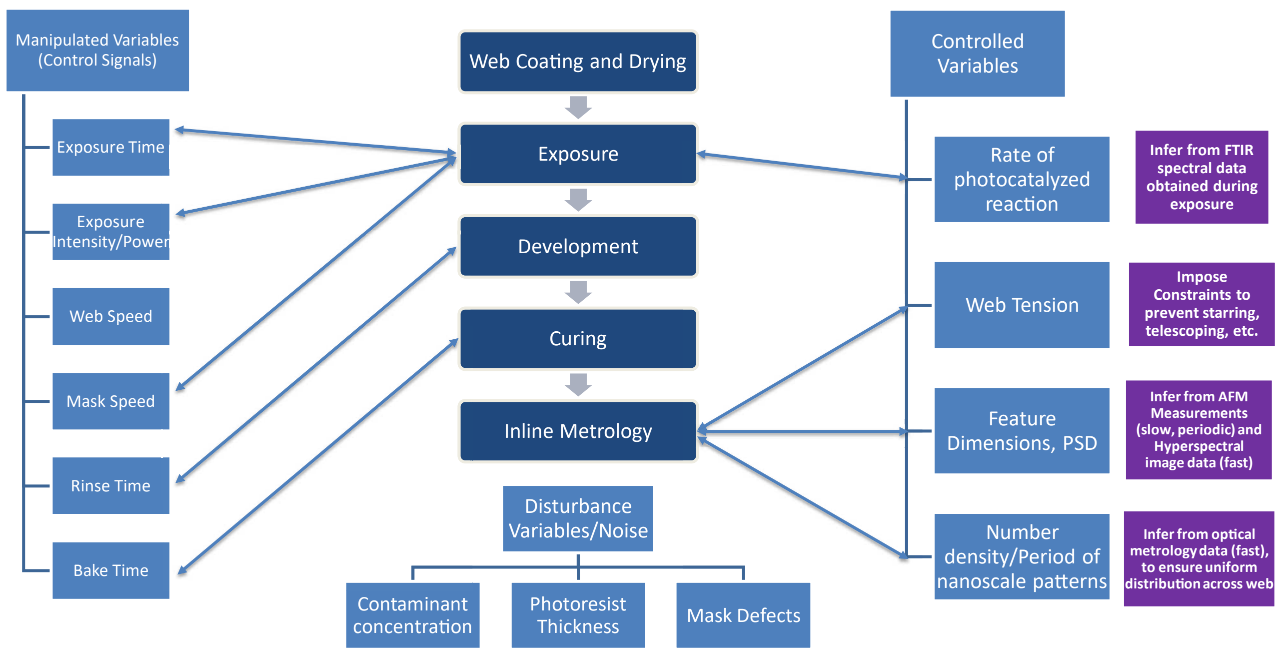

Figure 4: Manipulated, controlled and disturbance variables for R2R nanomanufacturing using near-field holographic lithography: an exemplar process.

development. Virtual metrology models can be constructed for each zone using data (line scan images, web tension, speed, rates of reaction, etc.) corresponding to that zone.

– The sequential single-line nature of R2R nanomanufacturing makes it more vulnerable to large process downtimes since a single piece of faulty equipment would result in the entire process shutting down. In general, the overall throughput is expected to be limited by the slowest intermediate process step. Scheduling of manufacturing operations in such systems requires further research, which could make use of reinforcement learning (RL)-based approaches (Wang et al. 2021).
– Production of multiple types of nanostructures would require intervention in the form of updating processing conditions in various units. For instance, in the case of IL, this could mean changes in photomasks used, light dosage, exposure times, or development conditions. Semiconductor manufacturing typically allows for the use of recyclable "pilot" or test wafers during process changeovers for equipment qualification or stabilization of process performance, due to the batch nature of unit operations (Yugma et al. 2015). In the entirely continuous R2R manufacturing setup, such an arrangement is infeasible, which could result in higher scrap rates than desired.

5 Conclusions and outlook

High-throughput continuous nanomanufacturing offers numerous opportunities for machine learning based yield control. One of the most impactful application areas for ML in manufacturing is computer vision (CV)-based part inspection and process monitoring at multiple length scales. Utilizing cost-effective sensors like spectral cameras coupled with ML algorithms can enable high-throughput part inspection, as established by a wide body of literature in the past decade (Li et al. 2019; Macaulay and Shafiee 2022; Park et al. 2016; Rossi et al. 2021). The findings of this article highlight the pivotal role ML can play in speeding up process control to real-time operations. ML models can be trained as virtual sensors or soft sensors, estimating critical process variables and quality attributes in real-time from available sensor data, enabling tight process monitoring without relying solely on physical sensors that may have measurement delays.

ML offers an effective approach for boosting throughput in continuous nanomanufacturing processes by leveraging two key techniques: noise reduction and image super-resolution. Inherent noise in sensor data can be filtered by ML algorithms trained to recognize and remove these patterns, leading to cleaner data for real-time monitoring and control (Tian et al. 2020; Yu et al. 2019). Furthermore, ML can enable image super-resolution by analyzing imperfect measurement data and reconstructing a high-resolution image of the nanostructures (Wang et al. 2021; Yang et al. 2019). This would allow for measurements at increased web speeds without compromising measurement quality.

Additionally, ML algorithms can build predictive models mapping process parameters to current and future process behavior and product quality, which can then be leveraged for model predictive control strategies, anticipating future dynamics, and taking proactive corrective actions to maintain optimal operating conditions in real-time. The parallelization and scalability of ML algorithms across distributed computing infrastructure allow for handling large volumes of sensor data and performing computationally intensive modeling and control tasks in real-time. However, challenges around handling process constraints, dynamics, and multi-objective optimization need to be addressed for effective process control and optimization. Limitations such as interpretability issues, lack of robustness, and ability to generalize to arbitrary processing conditions necessitate a case-by-case cost-benefit analysis. Established algorithms with well-defined physical models and limited data needs may be preferable due to their lower computational complexity and explainable outputs. Other challenges include handling noise, data scarcity and correlating defects with process parameters. A collaborative approach, where ML complements established physics-based methods, is likely for the foreseeable future.

While parallel deployment of multiple ML models in continuous nanomanufacturing is natural, a parsimonious approach is recommended. Complex models can be computationally expensive on standard hardware, and excessive models hinder human intervention during troubleshooting. Data-driven discovery of underlying physics, leading to interpretable process models, is preferable over opaque "black box" models.

Reliable AI models for multi-scale, nonlinear processes require strategic sensor placement. Optimizing scanning paths for high-resolution data and collecting multi-zone optical measurements are crucial for capturing process behavior across different length scales. However, controlling variables across these scales can lead to numerical instability (model stiffness) for simulation and optimization owing to vast differences in characteristic scales. A major challenge lies in developing theoretical criteria for observability (state estimation) and controllability (state manipulation) in these systems, especially considering the nonlinearities inherent to neural network models. For multiphysics, multi-scale phenomena, sensor placement needs to account for intricate cross-scale interactions to effectively capture these complexities in a ML model. Furthermore, ensuring observability and controllability with opaque neural networks remains an obstacle for reliable sensor placement and model building. In essence, optimizing sensor placement for robust AI models in such domains necessitates interdisciplinary research addressing challenges in efficient data acquisition, multi-zone coverage, bridging disparate scales, mitigating numerical stiffness, and developing theoretical frameworks for observability and controllability analysis of complex ML models.

In conclusion, bridging the gap between the promise and reality of ML in nanomanufacturing requires a synergistic research approach. This demands the formation of collaborative teams comprised of specialists in disparate fields. Machine learning engineers will be crucial for developing robust algorithms and data pipelines. Nanotechnologists and materials scientists will provide critical domain expertise to guide model development and ensure physical feasibility. These steps would also help address data scarcity for developing ML models. Researchers should encourage data sharing, establish centralized repositories, and engage the broader scientific community to contribute anonymized datasets. By embedding domain-specific physical laws into ML models, we can enhance their accuracy and feasibility within the nanomanufacturing context.

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References

Alexander Liddle, J. and Gallatin, M.G. (2016). Nanomanufacturing: a perspective. *ACS Nano* 10: 2995–3014.

Anand, U., Ghosh, T., Aabdin, Z., Vrancken, N., Yan, H., Xu, X., Holsteyns, F., and Mirsaidov, U. (2021). Deep learning-based high throughput inspection in 3D nanofabrication and defect reversal in nanopillar arrays: implications for next generation transistors. *ACS Appl. Nano Mater.* 4: 2664–2672.

Astrom, K.J. (1967). Computer control of a paper machine—an application of linear stochastic control theory. *IBM J. Res. Dev*. 11: 389–405.

Baek, K.H., Edgar, T.F., Song, K., Choi, G., Cho, H.K., and Han, C. (2014). An effective procedure for sensor variable selection and utilization in plasma etching for semiconductor manufacturing. *Comput. Chem. Eng.* 61: 20–29.

Bagal, A., Zhang, X.A., Shahrin, R., Dandley, E.C., Zhao, J., Poblete, F.R., Oldham, C.J., Zhu, Y., Parsons, G.N., Bobko, C., et al. (2017). Large-area nanolattice film with enhanced modulus, hardness, and energy dissipation. *Sci. Rep.* 7, https://doi.org/10.1038/s41598-017-09521-6.

Baldea, M. and Harjunkoski, I. (2014). Integrated production scheduling and process control: a systematic review. *Comput. Chem. Eng.* 71: 377–390.

Baldea, M., Du, J., Park, J., and Harjunkoski, I. (2015). Integrated production scheduling and model predictive control of continuous processes. *AIChE J.* 61: 4179–4190.

Barkhordari, A., Mashayekhi, H.R., Amiri, P., Özçelik, S., Hanife, F., and Azizian-Kalandaragh, Y. (2024). Performance of machine learning algorithms in spectroscopic ellipsometry data analysis of ZnTiO3 nanocomposite. *Sci. Rep.* 14: 1617.

Bougie, N., Onishi, T., and Tsuruoka, Y. (2022). Local control is all you need: decentralizing and coordinating reinforcement learning for large-scale process control. In: *2022 61st annual conference of the society of instrument and control engineers (SICE)*. IEEE, pp. 468–474.

Brown, K.A., Brittman, S., Maccaferri, N., Jariwala, D., and Celano, U. (2020). Machine learning in nanoscience: big data at small scales. *Nano Lett.* 20: 2–10.

Bunday, B.D., Bello, A., Solecky, E., and Vaid, A. (2018). 7/5nm logic manufacturing capabilities and requirements of metrology. In: Adan, O. and Ukraintsev, V.A. (Eds.). *Proceedings volume 10585, Metrology, inspection, and process control for microlithography XXXII. Conference: SPIE advanced lithography 2018*. SPIE, San Jose, California, United States.

Cai, M. and Vasile, C.I. (2021). Safe-critical modular deep reinforcement learning with temporal logic through gaussian processes and control barrier functions. *arXiv preprint arXiv:2109.02791*.

Campbell, S.A. (2001). *The science and engineering of microelectronic fabrication*. Oxford Univ. Press, New York.

Campbell, M., Sharp, D.N., Harrison, M.T., Denning, R.G., and Turberfield, A.J. (2000). Fabrication of photonic crystals for the visible spectrum by holographic lithography. *Nature* 404: 53–56.

Caspari, A., Tsay, C., Mhamdi, A., Baldea, M., and Mitsos, A. (2020). The integration of scheduling and control: top-down vs. bottom-up. *J. Process Control* 91: 50–62.

Chang, C.H., Tian, L., Hesse, W.R., Gao, H., Choi, H.J., Kim, J.G., Siddiqui, M., and Barbastathis, G. (2011). From two-dimensional colloidal self-assembly to three-dimensional nanolithography. *Nano Lett.* 11: 2533–2537.

Chou, S.Y., Krauss, P.R., and Renstrom, P.J. (1996). Imprint lithography with 25-nanometer resolution. *Science* 272: 85–87.

Cizmar, P., Vladár, A.E., and Postek, M.T. (2011). Real-time scanning charged-particle microscope image composition with correction of drift. *Microsc. Microanal.* 17: 302–308.

Connolly, L.G., Yao, T.F., Chang, A., and Cullinan, M. (2019). A tip-based metrology framework for real-time process feedback of roll-to-roll fabricated nanopatterned structures. *Precis. Eng.* 57: 137–148.

Cumpston, B.H., Ananthavel, S.P., Barlow, S., Dyer, D.L., Ehrlich, J.E., Erskine, L.L., Heikal, A.A., Kuebler, S.M., Lee, I.-Y.S., McCord-Maughon, D., et al. (1999). Two-photon polymerization initiators for three-dimensional optical data storage and microfabrication. *Nature* 398: 51–54.

Dai, W., Li, T., Zhang, L., Jia, Y., and Yan, H. (2022). Multi-rate layered operational optimal control for large-scale industrial processes. *IEEE Trans. Ind. Inf.* 18: 4749–4761.

Dave, P., Willig, D.A., Kudva, G.K., Pekny, J.F., and Doyle, F.J. (1997). LP methods in MPC of large-scale systems: application to paper-machine CD control. *AIChE J.* 43: 1016–1031.

den Boef, A.J. (2016). Optical wafer metrology sensors for process-robust CD and overlay control in semiconductor device manufacturing. *Surf. Topogr. Metrol. Prop.* 4: 023001.

Denno, P., Dickerson, C., and Harding, J.A. (2018). Dynamic production system identification for smart manufacturing systems. *J. Manuf. Syst.* 48: 192–203.

Dou, N.G., Jagt, R.A., Portela, C.M., Greer, J.R., and Minnich, A.J. (2018). Ultralow thermal conductivity and mechanical resilience of architected nanolattices. *Nano Lett.* 18: 4755–4761.

Dreyfus, P.A., Psarommatis, F., May, G., and Kiritsis, D. (2022). Virtual metrology as an approach for product quality estimation in Industry 4.0: a systematic review and integrative conceptual framework. *Int. J. Prod. Res.* 60, https://doi.org/10.1080/00207543.2021.1976433.

Dumont, G.A., Jonsson, I.M., Davies, M.S., Ordubadi, F.T., Fu, Y., Natarajan, K., Lindeborg, C., and Heaven, E.M. (1993). Estimation of moisture variations on paper machines. *IEEE Trans. Control Syst. Technol.* 1: 101–113.

Evans, K.E. (1991). Auxetic polymers: a new range of materials. *Endeavour* 15: 170–174.

Fan, S.-K.S. and Chang, Y.-J. (2013). An integrated advanced process control framework using run-to-run control, virtual metrology and fault detection. *J. Process Control* 23: 933–942.

Fan, Z., Razavi, H., Do, J.W., Moriwaki, A., Ergen, O., Chueh, Y.L., Leu, P.W., Ho, J.C., Takahashi, T., Reichertz, L.A., et al. (2009). Three-dimensional nanopillar-array photovoltaics on low-cost and flexible substrates. *Nat. Mater.* 8: 648–653.

Faria-Briceno, J.J., Zhu, R., Sasidharan, V., Neumann, A., Singhal, S., Sreenivasan, S.V., and Brueck, S.R.J. (2019). Optical angular scatterometry: in-line metrology approach for roll-to-roll and nanoimprint fabrication. *J. Vac. Sci. Technol. B, Nanotechnol. Microelectron. Mater. Process. Meas. Phenom.* 37, https://doi.org/10.1116/1.5119707.

Feng, D., Wagner, R.B., and Raman, A. (2021). Measuring nonuniform web tension for roll-to-roll manufacturing of flexible and printed electronics. *Flex. Print. Electron.* 6: 035006.

Fourkas, J.T., Gao, J., Han, Z., Liu, H., Marmiroli, B., Naughton, M.J., Petersen, J.S., Sun, Y., Vagilio Pret, A., and Zheng, Y. (2021). Grand challenges in nanofabrication: there remains plenty of room at the bottom. *Front. Nanotechnol.* 3, https://doi.org/10.3389/fnano.2021.700849.

Ganesan, R., Das, T.K., and Ramachandran, K.M. (2007). A multiresolution analysis-assisted reinforcement learning approach to run-by-run control. *IEEE Trans. Autom. Sci. Eng.* 4, https://doi.org/10.1109/tase.2006.879915.

Gao, J., Li, P., Chen, Z., and Zhang, J. (2020). A survey on deep learning for multimodal data fusion. *Neural Comput.* 32: 829–864.

Gawlik, B., Barrera, C., Yu, E.T., and Sreenivasan, S.V. (2020). Hyperspectral imaging for high-throughput, spatially resolved spectroscopic scatterometry of silicon nanopillar arrays. *Opt. Express* 28: 14209.

Gawlik, B., Barr, A.R., Mallavarapu, A., and Sreenivasan, S.V. (2021). Spectral imaging and computer vision for high-Throughput defect detection and root-cause analysis of silicon nanopillar arrays. *J. Micro Nano-Manuf.* 9, https://doi.org/10.1115/1.4049959.

Gostimirovic, D., Grinberg, Y., Xu, D.-X., and Liboiron-Ladouceur, O. (2023). Improving fabrication fidelity of integrated nanophotonic devices using deep learning. *ACS Photon.* 10: 1953–1961.

Graff, N. and Djurdjanovic, D. (2022). Modelling, simulation and control of roll-to-roll physical vapor deposition processes. *Procedia CIRP* 113: 546–551.

Guo, T., Dong, J., Li, H., and Gao, Y. (2017). Simple convolutional neural network on image classification. In: *2017 IEEE 2nd international conference on big data analysis (ICBDA), Beijing, China*. IEEE, pp. 721–724.

He, N., Shi, D., Wang, J., Forbes, M., Backström, J., and Chen, T. (2015). User friendly robust MPC tuning of uncertain paper-making processes. In: *IFAC-PapersOnLine, Whistler, British Columbia, Canada*. IFAC, pp. 1021–1026.

Horgan, C.C., Jensen, M., Nagelkerke, A., St-Pierre, J.P., Vercauteren, T., Stevens, M.M., and Bergholt, M.S. (2021). High-throughput molecular imaging via deep-learning-enabled Raman spectroscopy. *Anal. Chem.* 93: 15850–15860.

Hussain, M., Bird, J.J., Faria, D.R. (2019). A study on CNN transfer learning for image classification. In: Lotfi, A., Bouchachia, H., Gegov, A., Langensiepen, C., McGinnity, M. (Eds.). *Advances in computational intelligence systems. UKCI 2018*. Advances in intelligent systems and computing, Vol. DCCCXL. Cham, Springer.

Hyun Baek, K., Song, K., Han, C., Choi, G., Ku Cho, H., and Edgar, T.F. (2014). Implementation of a robust virtual metrology for plasma etching through effective variable selection and recursive update technology. *J. Vac. Sci. Technol. B, Nanotechnol. Microelectron. Mater. Process. Meas. Phenom.* 32, https://doi.org/10.1116/1.4862254.

Jaeger, C.J. (2002). *Introduction to microelectronic fabrication*. Prentice Hall, New Jersey.

Jang, D., Meza, L.R., Greer, F., and Greer, J.R. (2013). Fabrication and deformation of three-dimensional hollow ceramic nanostructures. *Nat. Mater.* 12: 893–898.

Jin, X., Shui, H., and Shpitalni, M. (2019). Virtual sensing and virtual metrology for spatial error monitoring of roll-to-roll manufacturing systems. *CIRP Ann.* 68: 491–494.

Kagias, M., Lee, S., Friedman, A.C., Zheng, T., Veysset, D., Faraon, A., and Greer, J.R. (2023). Metasurface-enabled holographic lithography for impact-absorbing nano-architected sheets. *Adv. Mater.*: 2209153, https://doi.org/10.1002/adma.202209153.

Kalinin, S.V., Ziatdinov, M., Hinkle, J., Jesse, S., Ghosh, A., Kelley, K.P., Lupini, A.R., Sumpter, B.G., and Vasudevan, R.K. (2021). Automated and autonomous experiments in electron and scanning probe microscopy. *ACS Nano* 15: 12604–12627.

Kang, S. and Kang, P. (2017). An intelligent virtual metrology system with adaptive update for semiconductor manufacturing. *J. Process Control* 52: 66–74.

Kang, P., Lee, H., Cho, S., Kim, D., Park, J., Park, C.-K., and Doh, S. (2009). A virtual metrology system for semiconductor manufacturing. *Expert Syst. Appl.* 36: 12554–12561.

Kang, P., Kim, D., Lee, H., Doh, S., and Cho, S. (2011). Virtual metrology for run-to-run control in semiconductor manufacturing. *Expert Syst. Appl.* 38: 2508–2522.

Kang, S., Park, J., and Lee, M. (2023). Machine learning-enabled autonomous operation for atomic force microscopes. *Rev. Sci. Instrum.* 94, https://doi.org/10.1063/5.0172682.

Khaleghi, B., Khamis, A., Karray, F.O., and Razavi, S.N. (2013). Multisensor data fusion: a review of the state-of-the-art. *Inf. Fusion* 14: 28–44.

Khan, A.A., Moyne, J.R., and Tilbury, D.M. (2007). An approach for factory-wide control utilizing virtual metrology. *IEEE Trans. Semicond. Manuf.* 20: 364–375.

Kim, Y., Hong, K., and Bang, H. (2018). Utilizing out-of-sequence measurement for ambiguous update in particle filtering. *IEEE Trans. Aero. Electron. Syst.* 54: 493–501.

Krauss, T.F., Rue, R.M.D. La, and Brand, S. (1996). Two-dimensional photonic-bandgap structures operating at near-infrared wavelengths. *Nature* 383: 699–702.

Kruit, P., Hobbs, R.G., Kim, C.-S., Yang, Y., Manfrinato, V.R., Hammer, J., Thomas, S., Weber, P., Klopfer, B., Kohstall, C., et al. (2016). Designs for a quantum electron microscope. *Ultramicroscopy* 164: 31–45.

Kumar, M., Garg, D.P., and Zachery, R.A. (2006). A generalized approach for inconsistency detection in data fusion from multiple sensors. In: *2006 American control conference, Minneapolis, MN, USA*. IEEE. https://doi.org/10.1109/ACC.2006.1656526.

Kumar, N., Kennedy, K., Gildersleeve, K., Abelson, R., Mastrangelo, C.M., and Montgomery, D.C. (2006). A review of yield modelling techniques for semiconductor manufacturing. *Int. J. Prod. Res.* 44: 5019–5036.

Lakes, R. (1987). Foam structures with a negative Poisson's ratio. *Science* 235: 1038–1040.

Lakes, R.S., Lee, T., Bersie, A., and Wang, Y.C. (2001). Extreme damping in composite materials with negative-stiffness inclusions. *Nature* 410: 565–567.

Lazar, A. and Fodor, P.S. (2015). Computational imaging XIII. In: Bouman, C.A. and Sauer, K.D. (Eds.). *Sparsity based noise removal from low dose scanning electron microscopy images*, 9401. SPIE, pp. 29–35.

Lee, J., Byeon, J., and Lee, C. (2020). Theories and control technologies for web handling in the roll-to-roll manufacturing process. *Int. J. Precis. Eng. Manufacturing-Green Technol.* 7: 525–544.

Lee, J., Shin, K., and Jung, H. (2022). Control scheme for rapidly responding register controller using response acceleration input in industrial roll-to-roll manufacturing systems. *IEEE Trans. Ind. Electron.* 69: 5215–5224.

Lee, J.H., Wang, L., Kooi, S., Boyce, M.C., and Thomas, E.L. (2010). Enhanced energy dissipation in periodic epoxy nanoframes. *Nano Lett.* 10: 2592–2597.

Leinen, P., Esders, M., Schütt, K.T., Wagner, C., Müller, K.R., and Stefan Tautz, F. (2020). Autonomous robotic nanofabrication with reinforcement learning. *Sci. Adv.* 6, https://doi.org/10.1126/sciadv.abb6987.

Li, J., Zhou, Q., Cao, L., Wang, Y., and Hu, J. (2022). A convolutional neural network-based multi-sensor fusion approach for in-situ quality monitoring of selective laser melting. *J. Manuf. Syst.* 64: 429–442.

Li, Q., Cai, W., Wang, X., Zhou, Y., Feng, D.D., and Chen, M. (2014). Medical image classification with convolutional neural network. In: *2014 13th international conference on control automation robotics & vision (ICARCV), Singapore*. IEEE, pp. 844–848.

Li, S., Song, W., Fang, L., Chen, Y., Ghamisi, P., and Benediktsson, J.A. (2019). Deep learning for hyperspectral image classification: an overview. *IEEE Trans. Geosci. Rem. Sens.* 57: 6690–6709.

Li, Y., Wu, Y., Yu, H., Takeuchi, I., and Jaramillo, R. (2021). Deep learning for rapid analysis of spectroscopic ellipsometry data. *Adv. Photon. Res.* 2, https://doi.org/10.1002/adpr.202100147.

Li, Z., Xue, S.-R., Yu, X.-H., and Gao, H.-J. (2020). Controller optimization for multirate systems based on reinforcement learning. *Int. J. Autom. Comput.* 17: 417–427.

Li, Z., Fan, Q., and Yin, Y. (2022). Colloidal self-assembly approaches to smart nanostructured materials. *Chem. Rev.* 122: 4976–5067.

Lin, S.Y., Fleming, J.G., Hetherington, D.L., Smith, B.K., Biswas, R., Ho, K.M., Sigalas, M.M., Zubrzycki, W., Kurtz, S.R., and Bur, J. (1998). A three-dimensional photonic crystal operating at infrared wavelengths. *Nature* 394: 251–253.

Liu, J., Zhang, D., Yu, D., Ren, M., and Xu, J. (2021). Machine learning powered ellipsometry. *Light: Sci. Appl.* 10: 55.

Liu, Shuo, Chen, X., Yang, T., Guo, C., Zhang, J., Ma, J., Chen, C., Wang, C., Zhang, C., and Liu, Shiyuan. (2022). Machine learning aided solution to the inverse problem in optical scatterometry. *Measurement* 191: 110811.

Liu, T., de Haan, K., Rivenson, Y., Wei, Z., Zeng, X., Zhang, Y., and Ozcan, A. (2019). Deep learning-based super-resolution in coherent imaging systems. *Sci. Rep.* 9: 3926.

Lu, S. and Jayaraman, A. (2023). Pair-variational autoencoders for linking and cross-reconstruction of characterization data from complementary structural characterization techniques. *JACS Au* 3: 2510–2521.

Lu, B., Stuber, J., and Edgar, T.F. (2014). Integrated online virtual metrology and Fault Detection in plasma etch tools. *Ind. Eng. Chem. Res.* 53: 5172–5181.

Lucas, A., Iliadis, M., Molina, R., and Katsaggelos, A.K. (2018). Using deep neural networks for inverse problems in imaging: beyond analytical methods. *IEEE Signal Process. Mag.* 35: 20–36.

Macaulay, M.O. and Shafiee, M. (2022). Machine learning techniques for robotic and autonomous inspection of mechanical systems and civil infrastructure. *Autonom. Intell. Syst.* 2: 8.

Mack, C.A. and Bunday, B.D. (2018). CD-SEM algorithm optimization for line roughness metrology. In: Ukraintsev, V.A. (Eds.). *Proceedings Volume 10585, Metrology, Inspection, and Process Control for Microlithography XXXII. Conference: SPIE advanced lithography 2018*. SPIE, San Jose, California, USA.

Maggipinto, M., Beghi, A., McLoone, S., and Susto, G.A. (2019). DeepVM: a Deep Learning-based approach with automatic feature extraction for 2D input data Virtual Metrology. *J. Process Control* 84: 24–34.

Maize, K., Mi, Y., Cakmak, M., and Shakouri, A. (2023). Real-time metrology for roll-to-roll and advanced inline manufacturing: a review. *Adv. Mater. Technol.* 8, https://doi.org/10.1002/admt.202200173.

Manohar, K., Brunton, B.W., Kutz, J.N., and Brunton, S.L. (2018). Data-driven sparse sensor placement for reconstruction: demonstrating the benefits of exploiting known patterns. *IEEE Control Syst. Mag.* 38: 63–86.

Meza, L.R., Das, S., and Greer, J.R. Strong, lightweight, and recoverable three-dimensional ceramic nanolattices, Available at: https://www.science.org.

Milor, L. (2013). A survey of yield modeling and yield enhancement methods. *IEEE Trans. Semicond. Manuf.* 26: 196–213.

Ming, Y., Yin, H., and Li, Y. (2022). On the impact of spurious correlation for out-of-distribution detection, Available at: https://github.com/deeplearning-wisc/Spurious_OOD.

Mönch, L., Fowler, J.W., Dauzère-Pérès, S., Mason, S.J., and Rose, O. (2011). A survey of problems, solution techniques, and future challenges in scheduling semiconductor manufacturing operations. *J. Sched.* 14: 583–599.

Natinsky, E., Khan, R.M., Cullinan, M., and Dingreville, R. (2024). Reconstruction of high-resolution atomic force microscopy measurements from fast-scan data using a Noise2Noise algorithm. *Measurement* 227: 114263.

Nesse, T., Simonsen, I., and Holst, B. (2019). Nanometer-resolution mask lithography with matter waves: near-field binary holography. *Phys. Rev. Appl.* 11, https://doi.org/10.1103/physrevapplied.11.024009.

Noda, S., Tomoda, K., Yamamoto, N., and Chutinan, A. (2000). Full three-dimensional photonic bandgap crystals at near-infrared wavelengths. *Science* 289: 604–606.

Nwanna, E.C., Coetzee, R.A.M., and Jen, T.C. (2022). A numerical approach on the selection of the purge flow rate in an atomic layer deposition (ALD) process. *Phys. Fluids* 34, https://doi.org/10.1063/5.0088872.

Paik, S., Kim, G., Chang, S., Lee, S., Jin, D., Jeong, K.Y., Lee, I.S., Lee, J., Moon, H., Lee, J., et al. (2020). Near-field sub-diffraction photolithography with an elastomeric photomask. *Nat. Commun.* 11, https://doi.org/10.1038/s41467-020-14439-1.

Palavesam, N., Marin, S., Hemmetzberger, D., Landesberger, C., Bock, K., and Kutter, C. (2018). Roll-to-roll processing of film substrates for hybrid integrated flexible electronics. *Flex. Print. Electron.* 3, https://doi.org/10.1088/2058-8585/aaaa04.

Park, J.-K., Kwon, B.-K., Park, J.-H., and Kang, D.-J. (2016). Machine learning-based imaging system for surface defect inspection. *Int. J. Precis. Eng. Manufacturing-Green Technol.* 3: 303–310.

Peled, E., Amit, E., Lamhot, Y., Svizher, A., Klein, D., Marchelli, A., Volkovich, R., Yaziv, T., Cheng, A., Lee, H., et al. (2018). Spectral tunability for accuracy, robustness, and resilience. In: Ukraintsev, V.A. (Ed.). *Metrology, inspection, and process control for microlithography XXXII*, 10585. SPIE, pp. 163–169.

Phung, T.H., Gafurov, A.N., Kim, I., Kim, S.Y., Kim, K.M., and Lee, T.M. (2021). IoT device fabrication using roll-to-roll printing process. *Sci. Rep.* 11, https://doi.org/10.1038/s41598-021-99436-0.

Plummer, J.D., Deal, M.D., and Griffin, P.B. (2000). *Silicon VLSI technology*. Prentice-Hall, New Jersey.

Potočnik, T., Christopher, P.J., Mouthaan, R., Albrow-Owen, T., Burton, O.J., Jagadish, C., Tan, H.H., Wilkinson, T.D., Hofmann, S., Joyce, H.J., et al. (2022). Automated computer vision-enabled manufacturing of nanowire devices. *ACS Nano* 16: 18009–18017.

Qi, M., Lidorikis, E., Rakich, P.T., Johnson, S.G., Joannopoulos, J.D., Ippen, E.P., and Smith, H.I. (2004). A three-dimensional optical photonic crystal with designed point defects. *Nature* 429: 538–542.

Qin, S.J., Cherry, G., Good, R., Wang, J., and Harrison, C.A. (2006). Semiconductor manufacturing process control and monitoring: a fab-wide framework. *J. Process Control* 16: 179–191.

Raul, P.R. and Pagilla, P.R. (2015). Design and implementation of adaptive PI control schemes for web tension control in roll-to-roll (R2R) manufacturing. *ISA Trans.* 56: 276–287.

Rippon, L.D., Lu, Q., Forbes, M.G., Gopaluni, R.B., Loewen, P.D., and Backström, J.U. (2019). Machine direction adaptive control on a paper machine. *Ind. Eng. Chem. Res.* 58: 11452–11473.

Rossi, A., Moretti, M., and Senin, N. (2021). Layer inspection via digital imaging and machine learning for in-process monitoring of fused filament fabrication. *J. Manuf. Process.* 70: 438–451.

Russell, E.L., Chiang, L.H., and Braatz, R.D. (2000). Fault detection in industrial processes using canonical variate analysis and dynamic principal component analysis. *Chemom. Intell. Lab. Syst.* 51: 81–93.

Sabbagh, R., Stothert, A., Sreenivasan, S.V., and Djurdjanovic, D. (2023a). Optical metrology of critical dimensions in large-area nanostructure arrays with complex patterns. *J. Manuf. Sci. Eng.*145, https://doi.org/10.1115/1.4056923.

Sabbagh, R., Stothert, A., and Djurdjanovic, D. (2023b). Machine learning for rapid inference of critical dimensions in optical metrology of nanopatterned surfaces. *CIRP J. Manuf. Sci. Technol.* 47: 184–192.

Santander, O., Giannikopoulos, I., Stadtherr, M.A., and Baldea, M. (2023). An integrated stochastic deep learning–short-term production scheduling–optimal control framework for general batch processes. *Ind. Eng. Chem. Res.* 62: 2124–2137.

Sarin, S.C., Varadarajan, A., and Wang, L. (2011). A survey of dispatching rules for operational control in wafer fabrication. *Prod. Plann. Control* 22: 4–24.

Shi, Y., Han, L., Han, L., Chang, S., Hu, T., and Dancey, D. (2022). A latent encoder coupled generative adversarial network (LE-GAN) for efficient hyperspectral image super-resolution. *IEEE Trans. Geosci. Rem. Sens.* 60, https://doi.org/10.1109/tgrs.2022.3193441.

Shui, H., Jin, X., and Ni, J. (2019). Twofold variation propagation modeling and analysis for roll-to-roll manufacturing systems. *IEEE Trans. Autom. Sci. Eng.* 16: 599–612.

Smith, T.H. and Boning, D.S. (1997). Artificial neural network exponentially weighted moving average controller for semiconductor processes. *J. Vac. Sci. Technol. A: Vac. Surf. Film.* 15: 1377–1384.

Smith, A., Schilling, S., and Daoutidis, P. (2024). Automated characterization and monitoring of material shape using Riemannian geometry. *Comput. Chem. Eng.* 181, https://doi.org/10.1016/j.compchemeng.2023.108525.

Song, G., Wang, Y., and Tan, D.Q. (2022). A review of surface roughness impact on dielectric film properties. *IET Nanodielectrics* 5: 1–23.

Spielberg, S., Tulsyan, A., Lawrence, N.P., Loewen, P.D., and Bhushan Gopaluni, R. (2019). Toward self-driving processes: a deep reinforcement learning approach to control. *AIChE J.* 65, https://doi.org/10.1002/aic.16689.

Staniewicz, L. and Midgley, P.A. (2015). Machine learning as a tool for classifying electron tomographic reconstructions. *Adv. Struct. Chem. Imag.* 1: 9.

Stewart, Gregory Edward and Sc, M. (2000). *Two dimensional loop shaping controller design for paper machine cross-directional processes*, Doctoral dissertation. Vancouver, University of British Columbia.

Su, A.J., Jeng, J.C., Huang, H.P., Yu, C.C., Hung, S.Y., and Chao, C.K. (2007). Control relevant issues in semiconductor manufacturing: overview with some new results. *Control Eng. Pract.* 15: 1268–1279.

Su, R., Park, S.H., Li, Z., and McAlpine, M.C. (2019). 3D printed electronic materials and devices. In: *Robotic systems and autonomous platforms [online]*. Woodhead Publishing in Materials, pp. 309–334. https://doi.org/10.1016/B978-0-08-102260-3.00013-5.

Sun, B., Fan, M., Liu, G., Lv, M., Liu, S., He, M., Huang, K., and Yang, C. (2024). A dynamics-learning multirate estimation approach for the feeding condition perception of complex industry processes. *IEEE Trans. Cybern.* 54: 3286–3298.

Sunaoshi, T., Kaji, K., Orai, Y., Schamp, C.T., and Voelkl, E. (2016). STEM/SEM, chemical analysis, atomic resolution and surface imaging at ≤ 30 kV with No aberration correction for nanomaterials on graphene support. *Microsc. Microanal.* 22: 604–605.

Tang, W. and Daoutidis, P. (2021). Dissipativity learning control (DLC): theoretical foundations of input–output data-driven model-free control. *Syst. Control Lett.* 147: 104831.

Tian, C., Fei, L., Zheng, W., Xu, Y., Zuo, W., and Lin, C.-W. (2020). Deep learning on image denoising: an overview. *Neural Networks* 131: 251–275.

Tom, M., Yun, S., Wang, H., Ou, F., Orkoulas, G., and Christofides, P.D. (2022). Machine learning-based run-to-run control of a spatial thermal atomic layer etching reactor. *Comput. Chem. Eng.* 168: 108044.

Tsay, C. and Baldea, M. (2020). Integrating production scheduling and process control using latent variable dynamic models. *Control Eng. Pract.* 94: 104201.

Ulbrich, M. and Bloemen Waanders, B.van. (2018). An introduction to partial differential equations constrained optimization. *Optim. Eng.* 19: 515–520.

Valenzuela, M.A., Bentley, J.M., and Lorenz, R.D. (2003). Sensorless tension control in paper machines. *IEEE Trans. Ind. Appl.* 39: 294–304.

Venkatasubramanian, V., Rengaswamy, R., Yin, K., and Kavuri, S.N. (2003). A review of process fault detection and diagnosis. *Comput. Chem. Eng.* 27: 293–311.

Vieu, C., Carcenac, F., Pépin, A., Chen, Y., Mejias, M., Lebib, A., Manin-Ferlazzo, L., Couraud, L., and Launois, H. (2000). Electron beam lithography: resolution limits and applications. *Appl. Surf. Sci.* 164: 111–117.

Wallentin, J., Anttu, N., Asoli, D., Huffman, M., Åberg, I., Magnusson, M.H., Siefer, G., Fuss-Kailuweit, P., Dimroth, F., Witzigmann, B., et al. (2013). InP nanowire array solar cells achieving 13.8% efficiency by exceeding the ray optics limit. *Science* 339: 1057–1060.

Wang, L., Pan, Z., and Wang, J. (2021). A review of reinforcement learning based intelligent optimization for manufacturing scheduling. *Complex Syst. Model. Simulat.* 1: 257–270.

Wang, Z., Chen, J., and Hoi, S.C.H. (2021). Deep learning for image super-resolution: a survey. *IEEE Trans. Pattern Anal. Mach. Intell.* 43: 3365–3387.

Westerhuis, J.A., Gurden, S.P., and Smilde, A.K. (2000). Generalized contribution plots in multivariate statistical process monitoring. *Chemom. Intell. Lab. Syst.* 51: 95–114.

Willmann, J., Stocker, D., and Dörsam, E. (2014). Characteristics and evaluation criteria of substrate-based manufacturing. Is roll-to-roll the best solution for printed electronics? *Org. Electron.* 15: 1631–1640.

Xie, J., Zhang, D., Yan, X.-Q., Ren, M., Zhao, X., Liu, F., Sun, R., Li, X., Li, Z., Chen, S., et al. (2019). Optical properties of chemical vapor deposition-grown PtSe 2 characterized by spectroscopic ellipsometry. *2D Materials* 6: 035011.

Yan, J. and Du, X. (2020). Web tension and speed control in roll-to-roll systems. In: *Control theory in engineering [working title]*. IntechOpen, London. 10.5772/intechopen.88797.

Yang, W., Zhang, X., Tian, Y., Wang, W., Xue, J.-H., and Liao, Q. (2019). Deep learning for single image super-resolution: a brief review. *IEEE Trans. Multimed.* 21: 3106–3121.

Yu, S., Ma, J., and Wang, W. (2019). Deep learning for denoising. *Geophysics* 84: V333–V350.

Yue, H.H., Qin, S.J., Markle, R.J., Nauert, C., and Gatto, M. (2000). *Fault detection of plasma etchers using optical emission spectra*. In: *IEEE Trans. Semicond. Manuf*., 13: 374–385.

Yue, X., Wang, K., Yan, H., Park, J.G., Liang, Z., Zhang, C., Wang, B., and Shi, J. (2017). Generalized wavelet shrinkage of inline Raman spectroscopy for quality monitoring of continuous manufacturing of carbon nanotube buckypaper. *IEEE Trans. Autom. Sci. Eng.* 14: 196–207.

Yue, X., Yan, H., Park, J.G., Liang, Z., and Shi, J. (2018). A wavelet-based penalized mixed-effects decomposition for multichannel profile detection of in-line Raman spectroscopy. *IEEE Trans. Autom. Sci. Eng.* 15: 1258–1271.

Yue, X., Park, J.G., Liang, Z., and Shi, J. (2020). Tensor mixed effects model with application to nanomanufacturing inspection. *Technometrics* 62: 116–129.

Yugma, C., Blue, J., Dauzère-Pérès, S., and Obeid, A. (2015). Integration of scheduling and advanced process control in semiconductor manufacturing: review and outlook. *J. Schedul.* 18: 195–205.

Yun, S., Tom, M., Ou, F., Orkoulas, G., and Christofides, P.D. (2022). Multivariable run-to-run control of thermal atomic layer etching of aluminum oxide thin films. *Chem. Eng. Res. Des.* 182: 1–12.

Zhang, H., Yu, X., and Braun, P.V. (2011). Three-dimensional bicontinuous ultrafast-charge and-discharge bulk battery electrodes. *Nat. Nanotechnol.* 6: 277–281.

Zhang, Q., Lu, S., Li, J., Li, D., Lu, X., Zhong, L., and Tian, J. (2021). Phase-shifting interferometry from single frame in-line interferogram using deep learning phase-shifting technology. *Opt. Commun.* 498, https://doi.org/10.1016/j.optcom.2021.127226.

Zhang, X., Hong, M., Dhople, S., and Elia, N. (2022). A stochastic multi-rate control framework for modeling distributed optimization algorithms. In: *International conference on machine learning*. PMLR, pp. 26206–26222.

Zhang, X.A., Chen, Y.-A., Bagal, A., and Chang, C.-H. (2017). Enhanced total internal reflection using low-index nanolattice materials. *Opt. Lett.* 42: 4123.

Zhang, X.A., Bagal, A., Dandley, E.C., Zhao, J., Oldham, C.J., Wu, B.I., Parsons, G.N., and Chang, C.H. (2015). Ordered 3D thin-shell nanolattice materials with near-unity refractive indices. *Adv. Funct. Mater.* 25: 6644–6649.

Zhang, X.A., Elek, J., and Chang, C.H. (2013). Three-dimensional nanolithography using light scattering from colloidal particles. *ACS Nano* 7: 6212–6218.

Zhao, F., Zhang, C., and Geng, B. (2024). Deep multimodal data fusion. *ACM Comput. Surv.* 56: 1–36.

Zheng, L., Zywietz, U., Birr, T., Duderstadt, M., Overmeyer, L., Roth, B., and Reinhardt, C. (2021). UV-LED projection photolithography for high-resolution functional photonic components. *Microsyst. Nanoeng.* 7, https://doi.org/10.1038/s41378-021-00286-7.

Zhu, P., Zhang, D., Niu, X., Liu, J., Ren, M., and Xu, J. (2024). A lightweight neural network for spectroscopic ellipsometry analysis. *Adv. Opt. Mater.* 12, https://doi.org/10.1002/adom.202301381.

Zou, M., Ma, Y., Yuan, X., Hu, Y., Liu, J., and Jin, Z. (2018). Flexible devices: from materials, architectures to applications. *J. Semicond.* 39, https://doi.org/10.1088/1674-4926/39/1/011010.