A Review of Statistical Methods for Quality Improvement and Control in Nanotechnology

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Nanotechnology has received a considerable amount of attention from various fields and has become a multidisciplinary subject, where several research ventures have taken place in recent years. This field is expected to affect every sector of our economy and daily life in the near future. Besides advances in physics, chemistry, biology, and other science-based technologies, the use of statistical methods has also helped the rapid development of nanotechnology in terms of data collection, treatment-effect estimation, hypothesis testing, and quality control. This paper reviews some instances where statistical methods have been used in nanoscale applications. Topics include experimental design, uncertainty modeling, process optimization and monitoring, and areas for future research efforts.

Key Words: Automatic Control; Experimental Design; Nanomanufacturing; Physical-Statistical Modeling; Statistical Quality Control; Stochastic Modeling.

"Nanotechnology is the understanding and control of matter at dimensions of roughly 1 to 100 nanometers (nms, [a nanometer equals 10^{-9} meters]), where unique phenomena enable novel applications. Encompassing nanoscale science, engineering, and technology, nanotechnology involves imaging, measuring, modeling, and manipulating matter at this length scale. At the nanoscale, the physical, chemical, and biological properties of materials differ in fundamental and valuable ways from the proper-

ties of individual atoms and molecules or bulk matter. Nanotechnology R&D is directed toward understanding and creating improved materials, devices, and systems that exploit these new properties." (National Nanotechnology Initiative (2008)).

In the near future, it is expected that nanotechnology will impact every sector of our economy and daily life. Roco (2004) characterized the development of nanotechnology into four generations, each with different featured products. The first generation, starting from about 2001, is characterized by *passive nanostructures*; the major focus of this generation is on nanostructured materials and tools for measurement and control of nanoscale processes. Popular examples include nanoparticle synthesis and processing, nanocoating, various catalysis, etc. The second generation is characterized by *active nanostructures*, according to Roco (2004). The research focus moves from nanostructured materials to novel devices and

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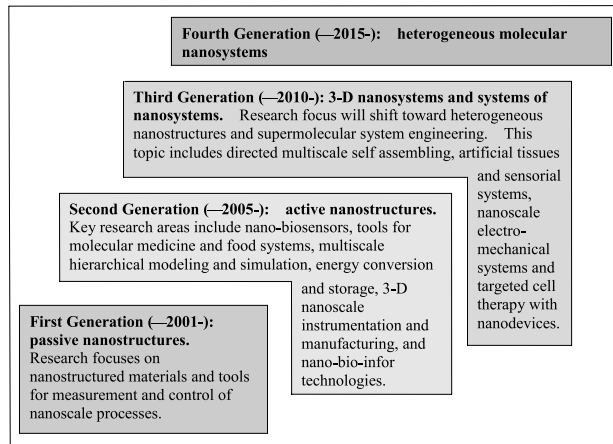
FIGURE 1. Four Generations of Nanotechnology Applications.

device system architectures. Popular research topics include nanobiosensors and nanodevices, nanoscale tools, nanoscale instrumentation, and nanomanufacturing. Modeling and simulation of nanoprocesses are also important topics in this generation. The third generation, featuring *3-D nanosystems and systems of nanosystems*, is expected to shift toward heterogeneous nanostructures and supermolecular system engineering. As an example, research on nanoscale electromechanical systems will be greatly promoted in this stage. Roco (2004) selected *heterogeneous molecular nanosystems* as the feature products of the fourth generation.

Figure 1 presents the overlapping generations of nanotechnology products and their respective manufacturing methods and research focus identified by the International Risk Governance Council (IRGC) (see Roco (2004) for details). We will use this summary to lead the discussion of where statistical methods can contribute (or have contributed) in nanotechnology development in the remainder of this paper.

Various government agencies, private corporations, and venture capitalists have created programs to support R&D in nanotechnology. According to Lux Research, the R&D investments in nanotechnology worldwide amounted to $12.4 billion in 2006. The United States Department of Energy started a nanotechnology program to develop new materials for improving fuel efficiency and providing efficient lighting. Likewise, the Department of Defense has laid out a research plan to develop new approaches and processes for manufacturing novel, reliable, lower cost, higher performance, and more flexible elec-

tronic, magnetic, optical, and mechanical devices. Even the research topics from the National Institutes of Health include new nanomaterials for interfacing with living tissues and molecular and cellular sensing for gathering diagnostic data inside human bodies where traditional instruments cannot reach.

Many challenges confronting nanotechnology research call for solutions from a multidisciplinary approach. Because statistical techniques have made sizable impacts in many technology fields in the past, statistics are expected to play an important role in tackling these challenges and boosting the development of nanotechnology. The purpose of this paper is to present examples of applications of statistical methods in nanotechnology research and to identify possible new statistical approaches for solving emerging problems in nanotechnology. Specifically, we highlight the following challenges that provide opportunities for statistical applications:

First, *statistical procedures help us learn more about the formation processes of nanocomposites*. Recently, researchers have reported new progress in developing nanocomposites. Forming processes dictate properties of nanocomposites. These processes usually involve complex chemical and mechanical reactions, where even minor changes of environmental factors or process settings may result in unexpected outcomes. Because the theory of the nanocomposite-forming process has not matured yet, many developments rely on experimental studies. Due to the high costs associated with experimental runs and sample measurements and the complex relationship between process outcomes and controllable/noise factors, new statistical experimental design methods are needed. Section 2 reviews recent work on applying experimental designs in nanocomposites applications and categorizes them based on the types of designs adopted in each study.

Second, *statistical modeling helps deal with special data types and processes*. Data collected from processes for developing nanoscale materials sometimes exhibit forms that are different from the usual patterns seen in conventional manufacturing situations. Such characteristics may include experimental spaces with many isolated regions of low or zero yields and high-frequency signals shown in spatial domains. Therefore, extensions of commonly used statistical techniques are required to draw useful information from such types of data. Beyond these extensions, stochastic modeling of the forming processes for characterizing multilevel or multiscale uncertain-

ties is another valuable topic. Section 3 summarizes recent work on data collection, statistical analysis, and uncertainty modeling in nano research and provides examples for researchers that are facing similar problems.

Third, *statistics help improve low-quality and high-defect processes*. Because nanosyntheses and nanofabrications are not well controlled thus far, defect rates of nanocomposites remain rather high. Mass and high-yield production is the key step confronting nanomanufacturing research. Statistical quality control and productivity improvement techniques, which proved to be tremendously helpful in traditional manufacturing, are needed in nanomanufacturing to enhance product quality and improve production efficiency. Extension of the traditional quality monitoring and improvement tools to accommodate the potential flood of sensor information is an interesting statistics research topic. Section 4 reviews the use of statistical process control (SPC) and automatic process control (APC) techniques in nanoapplications. The feedback control section provides examples of using stochastic partial differential equations (SPDEs) to describe thin-film deposition processes. Based on these SPDEs and results in kinetic Monte-Carlo simulations, multiscale automatic process controllers (APCs) are developed.

Last, *statistics help improve low and unpredictable product reliability*. Reliability models for nanocomposites are not well developed. However, through investigation of nanomaterials interfacing with environmental and stress factors, the reliability characteristics of nanodevices can be better understood and predicted. Jeng et al. (2007) provide a comprehensive review on recent reliability studies in nanoapplica-

tions. To keep this article brief, reliability issues will not be discussed in this review.

The remainder of this paper is organized as follows. Section 2 reviews successful applications of design of experiments (DOEs) techniques in nanotechnology research, which covers regular, nonregular, and robust parameter designs. Section 3 reviews various data collection, statistical analysis, and modeling methods in the literature. Specifically, works on probability distribution and variation modeling of nanostructure characteristics, treatment of high-frequency signal and spatial data as well as stochastic modeling techniques are presented. Section 4 reviews recent development of SPC and quality-control techniques. Examples of process monitoring and feedback control, especially multiscale modeling and control techniques, are illustrated in this section. Section 5 concludes this review with suggestions for future research on quality improvement and control in nanotechnology.

Design of Experiments

Due to the applications in nanoelectronics, photonics, data storage, and sensing, synthesizing nanostructures is a research topic of foremost importance in nanotechnology (Dasgupta et al. (2008)). However, the synthesis process is extremely sensitive to control settings and environmental noises. For example, to generate the nanostructures of nanosaws, nanowires, and nanobelts shown in Figure 2, Dasgupta et al. (2008) shows that the control factors, such as temperature and pressure, have a heavy impact on the final output of a synthesis process. Moreover, situations occur in which multiple responses are studied with functional relationships containing
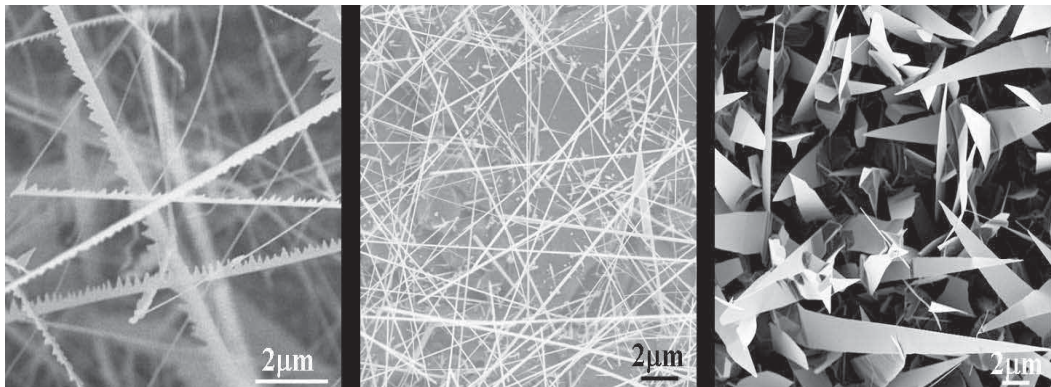

FIGURE 2. Nanosaws, Nanowires, and Nanobelts (from Dasgupta et al. (2008)).

many potential factors of interest. In order to robustly optimize several properties of nanoproducts, the impact of each factor, plus their interactions, on process outputs should be studied using advanced statistical techniques. In the literature, DOE has been employed as the major tool for exploring the relationship between controllable/noise factors and process responses. Regular designs, including full factorial and fractional factorial designs, response surface methodologies, and nonregular designs (e.g., D-optimal designs) have been successfully utilized to optimize synthesis processes and investigate material properties. This section reviews the use of DOE techniques and robust parameter designs in nanotechnology and gives examples of how these techniques can be utilized in the nanomanufacturing processes. This review covers studies in the following journals: *AAPS PharmSciTech*; *Carbon*; *Chemical Engineering Journal*, *Colloids and Surfaces A: Physicochemical Engineering Aspects*; *International Journal of Pharmaceutics*; *Journal of Physical Chemistry B*; *Journal of the American Statistical Association*; *Materials and Design*; *Microelectronics Reliability*; *Nanotechnology*; and *Powder Technology*.

Regular Designs

Basumallick et al. (2003) investigated the synthesis processes for Ni–SiO$_2$ and Co–SiO$_2$ nanocomposites, of which the physical properties are very sensitive to process parameters. This attribute makes the response surface very rugged. Because processing conditions have significant impact on the properties of nanocomposites, the authors considered three factors with three levels each in their experimentation. Instead of using a conventional three-level fractional factorial design, the authors designed eight runs using a two-level full factorial design and added three runs setting all factors at the middle level. Using regression equations fitted by the 11-run experiment outcomes, the fractional conversion values of the nanocomposites were modeled as a function of heating rate, composite concentration, and the starting temperature of the reaction. Because the response surface is very rugged, in order to improve the data collection and analysis methods used in this paper, we recommend the experimental designs (and their extensions) commonly used in computer experiments (e.g., Dasgupta et al. (2008)) for efficiently collecting costly data. Moreover, robust process-optimization ideas (e.g., Taguchi (1986)) could be used so that the physical properties of the nanocomposites are less sensitive to the noise factors.

Lin et al. (2003) studied the surface and grain structure of silver-plated film on a copper lead frame. The surface roughness was measured by atomic force microscopy (AFM), while the surface thickness was obtained by nanoindentation measurement using a UMIS-2000 nanoindenter. Additionally, the grain structure was measured by a transmission electron microscope (TEM). The impact of the silver-plated film-surface characteristics and grain structures to the quality of wedge bonding between gold wire and silver-plated lead frame were investigated through a 2^4 factorial design. Four design parameters were bond time, bond force, electrical current, and the two lead frame types. The experimental results showed that the surface characteristics and grain structures have a great impact on the quality of bonding.

To systematically obtain a model of factors leading to an optimum response, many researchers use the response surface methodology (RSM) for data collection and process optimization. RSM should also be considered when complex models are needed for characterizing a nanosynthesis or nanomanufacturing process. Prakobvaitayakit and Nimmannit (2003) presented a process to prepare polylactic-co-glycolic acid (PLGA) nanoparticles by interfacial deposition following the solvent-displacement technique. They investigated the process through a 2^3 factorial design experiment. Multiple responses—particle size, amount of encapsulated material, and encapsulation efficiency—were considered in the experiments. To optimize multiple responses, the authors used a simultaneous optimization technique with a desirability function. The desirability function converts each response into an individual function that varies over the range [0, 1] and takes the value one when the response is at its target value and less than one if not (see page 425 in Montgomery (2005) for details). By defining the overall desirability as the product of all individual functions, optimizing multiple responses is achieved by maximizing the overall desirability. With this method, the experimental design helped choose the optimal formulation ingredients for the nanoparticles. Now, the composites formed by PLGA nanoparticles are being commercially used for drug-delivery systems.

Barglik-Chory et al. (2004) used a second-order response surface to study the main effects and interactions of three controllable factors in synthesizing a type of semiconductor nanoparticles, colloidal CdS. The quantum effects in nanoparticles provided discrete energy levels, and the semiconductor band gap exhibited strong size dependence. Through con-

trolled experiments, the influence of environmental factors on nanoparticles could be investigated and quantified, potentially leading to an improvement in the nanofabrication efficiency.

After identifying the significant parameters by using Taguchi's parameter design method (Taguchi (1986)), Hou et al. (2007) used RSM to build a relationship between five process parameters and average grain size of nanoparticles. They found that the relationship between process parameters and grain size can be modeled with a second-order equation. Using the integrated genetic algorithm and RSM approach, the optimal settings of these five parameters in the nanoparticle milling process were determined.

Yördem et al. (2008) used RSM to investigate effects of material choices and process parameters on the diameter of electrospun polyacrylonitrile nanofibers. Three explanatory factors—voltage, solution concentration, and collector distance—and two response variables—fiber diameter and coefficient of variation—were studied. At each level of the collector distance, the other two factors were varied and the resultant fiber diameter was recorded. For the purpose of predicting fiber diameter and coefficient of variation, polynomial regression models were fitted to the experimental data at each level of the collector distance. Interactions among the factors were also identified. The authors suggested a narrower window of factor space for future nanofiber production.

Nazzal et al. (2002) used a three-level Box–Behnken design to evaluate the effect of formulation ingredients on the release rate of ubiquinone from its adsorbing solid compact and to obtain an optimized self-nanoemulsified tablet formulation. To study the effects of three independent variables on six responses, 15 runs of experiments were conducted. The formulation ingredients—copolyvidone, maltodextrin, and microcrystalline cellulose—were shown to have a significant effect on the emulsion release rate. However, this article did not show how to allocate these ingredients to optimize the multiple objectives.

Using a radio-frequency plasma reactor, Cota-Sanchez et al. (2005) used a 2^4 factorial design to study fullerenes synthesis. The response variable was C_{60} yield and the four operating parameters were reactor pressure, raw-material feed rate, carbon black–catalyst ratio, and generator-plate power. Using ultraviolet spectrometric analysis to measure yield, they found that the significant factors that affect the C_{60} synthesis are the reactor pressure, plate power, and feed rate. Under the optimal operating condition, the yields were synthesized up to about 7.7 wt.%; moreover, nanotubes were successfully produced. Further optimization of other parameters, such as the particle injection and quenching conditions, through the use of RSM might lead to enhanced yields of these carbon nanostructures.

In order to study the flexural properties and dispersions of a ceramic material, SiC, Yong and Hahn (2005) employed a two-level full-factorial design to investigate interactions between coupling agent and dispersant. A central composite design was used to determine the optimal dosages of chosen factors to achieve the maximum flexural strength and maximum particle dispersion. Experimental results showed that two objectives could be optimized simultaneously at the same factor levels. When an optimal dosage is employed, the nanoparticle reinforcement can enhance the mechanical properties of the composites.

Compared with factorial designs, split-plot designs are suitable for situations in which physical restrictions on a process exist and certain combinations of factor levels are difficult to reach. Nembhard et al. (2006) presented a split-plot design for investigating nanoscale milling of submicron channels on a gold layer. Similar to a synthesis process that involves complex chemical and mechanical reactions, such a milling process was sensitive to various controllable and noise factors. By treating the experiment as a two-stage process, whole-plot and subplot factors were identified. Results showed that split-plot experiments in nanomanufacturing reduce labor and costs and were often effective at detecting the effects of subplot factors.

In order to achieve high yield and reproducibility of a synthesis process, Dasgupta et al. (2008) conducted a full-factorial experiment. Two parameters, temperature and pressure, with five and nine levels, respectively, were varied during the experiment and three response variables, the numbers of nanosaws, nanowires, and nanobelts, were recorded. Because the total number of the nanostructures was a constant, the authors proposed simultaneously modeling the probability of generating different nanostructures using a multinomial generalized linear model (GLM). The optimal settings obtained from this research led to a significant improvement in the process yield.

Nonregular Designs

When a regular factorial design is not feasible due to possible limitations in experimental run-size and factor-level selections, some nonregular designs may be considered for nano process modeling and optimization. Among others, successful applications of D-optimal designs in nanotechnology have been demonstrated by several researchers. Compared with the regular designs, D-optimal designs may use nonorthogonal design matrices to reduce experimental runs and minimize the variances of coefficients associated with a specific model setting.

Fasulo et al. (2004) studied the extrusion processing of thermoplastic olefin (TPO) nanocomposites. Properties of TPO composites are influenced by the forming process. The authors chose different combinations of processing factors, including melt temperature, feed rate, and extruder screw-rotation speed, and investigated both surface appearance and physical/mechanical properties of the nanocomposites. A D-optimal design was adopted in the research to characterize the relationship between the quality measures and explanatory factors. Useful suggestions for optimizing process output were obtained.

Dasgupta (2007) developed a sequential minimum energy design (SMED) to deal with complex response surface with multiple optima for the yield of nanomaterial synthesis by using fewer design points. Figure 3 illustrates the response surface of the yield and the selected design points. Compared with traditional designs, the SMED design can probe high-yield regions and avoid nonyield points more effectively.

Robust Parameter Design

One major challenge confronting nanosynthesis/manufacturing processes is the high variation in experimental results. Most synthesis processes are very sensitive to environmental or noise factors. Therefore, robust parameter design (e.g., Taguchi (1986)) has been considered by several researchers to reduce experimental variation and enhance process yield and production efficiency. Figure 4 illustrates one of the key ideas of the robust parameter design. The level setting of control factors will be different by including the noise factors in the experiment when there are interactions between control and noise factors.

Nanoparticles have been widely utilized in many industrial applications, such as carbon nanotube, nanoceramics, and nanocompound materials. The wet-type milling machine is a recently developed tool to produce nanoparticles and to avoid aggregation effect. Because of its simplicity and applicability to all classes of materials, this machine is becoming popular. Hou et al. (2007) applied the robust parameter design method to optimize a nanoparticle milling process. They considered the following five process factors, each with three levels, to improve the nanoparticle milling process: the milling time, flow velocity of circulation systems, rotation velocity of agitator shaft, solute-to-solvent weight ratio, and filling ratio of grinding media. The response variable was the nanoparticle grain size, which is measured by the Coulter multisizer equipment. To save experimental cost, the L_{27} orthogonal array was used. The experimental results showed that all five process factors significantly affect the grain size.

Kim et al. (2004) implemented the robust design method with an L9 orthogonal array to optimize the recipe for preparing nanosize silver particles. The silver nanoparticles have been widely used in chemical and medical industry due to their unique properties of conductivity and resistance to oxidation. The objective was to determine the experimental conditions where the size of nanoparticle is small and has less variability. The following three factors were considered in the experiment: molar concentration ratio, dispersant concentration, and feed rate. The response variables are the average size and the size distribution of silver nanoparticles. The concentration of dispersant was identified as the most influencing factor on the average particle size and the size distribution. Using the derived optimal conditions, silver nanoparticles can now be prepared with small size variance using the derived optimal condition.

In another paper about the synthesis of nanoparticles, Kim et al. (2005) used Taguchi's method to optimize a new microemulsion method for preparing TiO_2 nanoparticles. An L_8 orthogonal array was used as the design of experiment for the five factors: H_2O surfactant value, H_2O/TEOT value, ammonia concentration, feed rate, and reaction temperature. The derived optimal condition led to the least size variability in nanoparticles.

Data Collection, Statistical Analysis, and Physical–Chemical–Statistical Modeling

Because measurements with complicated data patterns are frequently seen in experiments in nanotechnology, the collection, analysis, and modeling

Uniform design

Minimum potential design

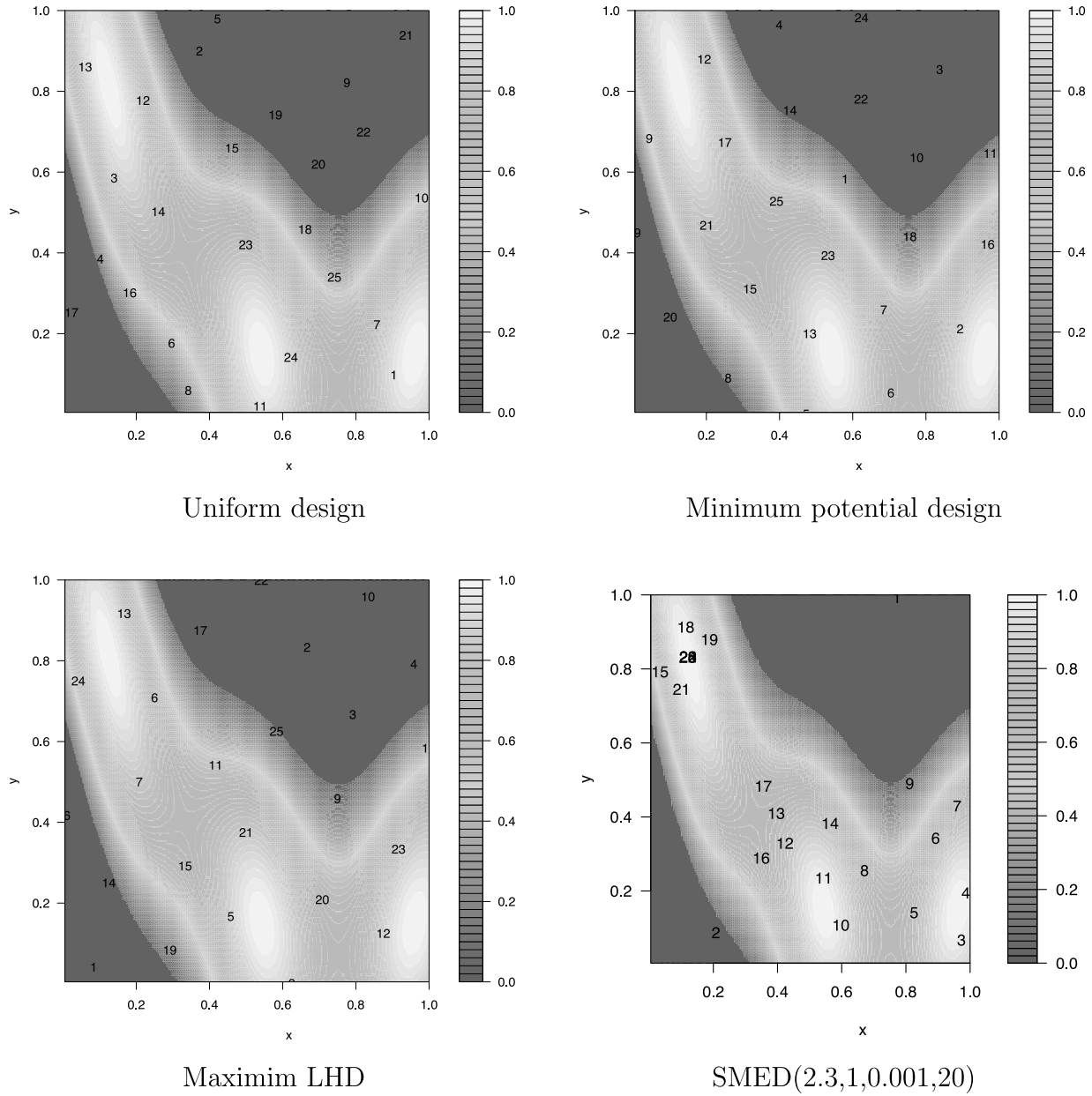Maximim LHD

SMED(2.3,1,0.001,20)

FIGURE 3. Response Surface of the Yield of a Nanostructure and the Selected Design Points (from Dasgupta (2007)).

of data in nanoengineering are vital topics. This section focuses on high-frequency and/or spatial data signals seen in nanotechnology studies. In addition, probability and stochastic models of nanoscale measurements and process variations are reviewed. The primary journals reviewed here include: *Combustion Theory and Modeling, IEEE Transactions on Nanotechnology, IEEE Transactions on Reliability, IEEE Transactions on Semiconductor Manufacturing, IEEE Transactions on Very Large Scale Integration (VLSI) Systems, IEICE Transactions on Electronics, International Journal of Engineering Science, International Journal of Plasticity, Journal of the Mechanics and Physics of Solids, Physical Review Letters, Solid-State Electronics*, and *Surface & Coatings Technology*.

Sampling Plans

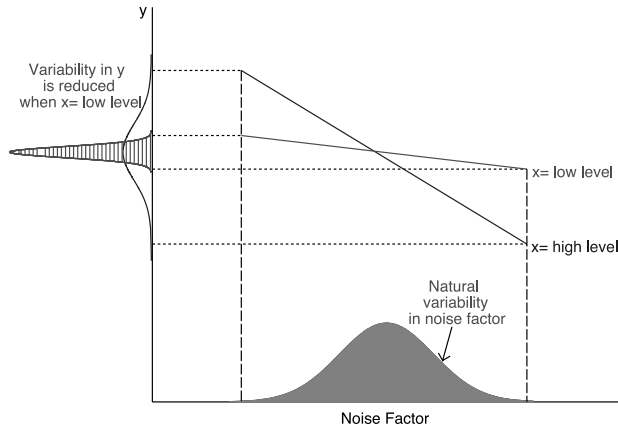Effective sampling plans are critical to model the nanofabrication processes using fewer data. The fol-

FIGURE 4. One of the Key Ideas of the Robust Parameter Design.

FIGURE 5. Idea Illustration of Importance Sampling.

lowing sampling techniques can be used to select representative data for analysis: simple random sampling, stratified sampling, cluster sampling, systematic sampling, importance sampling, and their hybrids. However, new sampling techniques have been developed for the nanofabrication process.

Zhao et al. (2004) developed a new double sampling technique to test interconnects and buses in very large scale integration (VLSI) circuits. Due to the rapid size scaling and high-speed operation of integrated circuits (ICs) using nanometer technologies, electromagnetic noise sources and their effects on interconnects have become extremely significant. The basic idea for this sampling technique was to sample test data for loading into two flip-flops at a fixed time interval and check whether the resultant data streams were consistent with each other. Thus, inconsistent and noisy signals with spikes could be captured. This technique was capable of detecting errors caused by electromagnetic noise effects in nanofabrication.

Chang et al. (2005) proposed a new method to detect the read failure in a static random access memory (SRAM) cell using a critical-point sampling technique. The idea was similar to the importance sampling. In sub–100-nm technologies, the analytical model previously used fails to accurately match realistic simulation results due to various short channel effects and different leakage components. It is preferable to employ a full-scale transient Monte Carlo (MC) simulation; however, using the MC method usually takes a large number of iterative runs to obtain an accurate read failure probability. Thus, rather than deriving the entire voltage-transfer characteris-
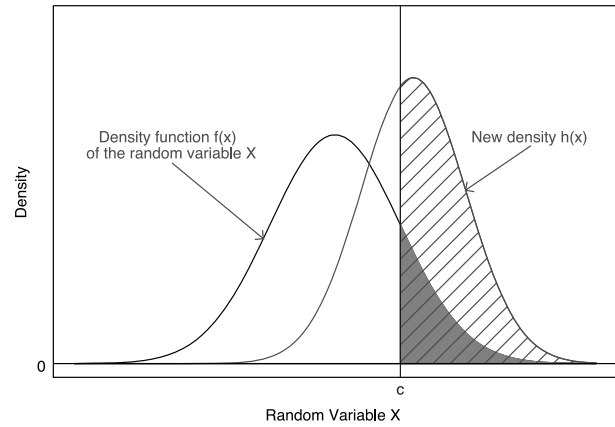
tic curve, the authors measured the SRAM cell stability at certain representative points on the curve for a specified voltage value. The experimental result showed that their model achieves high accuracy and was 20 times faster in computational speed.

Proper use of importance sampling will provide a more accurate inference or save the sampling cost with smaller sample size. This is a very helpful statistical technique for collecting expensive samples in many nanofabrication processes. Figure 5 illustrates the general idea. Suppose the target of the inference is the mean of a statistic $m(X)$, i.e. $E(m(X))$, where $m(x)$ only depends on the sample values x greater than a constant c. Then a proper choice of a new density $h(x)$ of X will make the sample mean of $m(X)f(X)/h(X)$ an unbiased estimator of the target with smaller variance. The main character of the new density $h(x)$ is to produce more samples on the area (greater than c) that affects the values of $m(x)$. See page 87 in Davison (2003) for more details of importance sampling.

Probability Distribution and Variation Modeling

As technology shrinks to the nanoscale region, probability distributions for material and structure performance may change to different forms. Moreover, variability in device performance becomes a major issue in the circuit-design stage. The following paragraphs review several papers studying probability distributions of nanoparticle measurements and process variations in IC manufacturing.

In ordnance technologies, the reproducibility of a

thermite's burning behavior is critical for both safety and performance evaluation. As a particle's diameter approaches the nanoscale, the burn-rate calculation becomes increasingly sensitive to variations in the particle diameter. In the study by Granier and Pantoya (2004), the burn-rate estimates for nanoscale thermites were statistically evaluated with a probability density function (pdf) of the particle-size distribution and a diameter-dependent burn-rate equation. Based on a series of scanning electron microscopy (SEM) images, a model of mass fractal aggregates was used to interpret the scattering data. A volume-weighted particle-size distribution was obtained. Both single mode and bimodal particle-size distributions were studied using Gaussian and a mixture of Gaussian distributions, respectively. The analysis showed that, as the particle size reduced to the nanoscale, the size distribution, rather than the average particle size alone, became increasingly important. Large variability in the burn rate was associated with a large standard deviation in particle sizes.

Bazant and Pang (2007) studied the pdf of strength of nanostructures based on a nanoscale atomic lattice. To predict a failure event with an extremely low probability, the cumulative distribution function (cdf) of strength of quasi-brittle structures was modeled as a chain of representative volume elements (RVE). Each of the RVEs was statistically characterized by a hierarchical model consisting of bundles (via a parallel coupling) of two long subchains. Each of the subchains consisted of subbundles of two or three long sub-subchains of sub-subbundles and so on. Eventually, the nanoscale atomic lattice was reached. They gave physical reasons that the failure of interatomic bonds follows a thermally activated process governed by stress-dependent activation energy barriers. The tail of the cdf of the RVE strength followed a power-law model. Finally, they concluded that the distribution of strength of the quasi-brittle nanostructure is a Weibull.

The physical process of surface formation in nanocomposites has several stochastic components. Chen et al. (2005) studied barrier effects impacting surface formation using oxidized porous silicon. According to their experimental results, a Gaussian distribution fit the data of oxidized porous silicon samples well. The barrier height turned out to have a Gaussion distribution, and the photon energy was shown to be a function of the barrier heights.

Hydrogenated amorphous silicon (a-Si:H), the prototypical disordered semiconductor, is an important material for use in nanoscale optoelectronic applications including sensors, flat-panel displays, 2D medical imaging, and photovoltaics. Belich et al. (2003) reported that the noise distribution in a-Si:H was non-Gaussian in nature, a surprising feature in macroscopic samples at and above room temperature. Traditionally, the noise arose from an ensemble of statistically independent fluctuators. This led to Gaussian-distributed noises. One possible explanation of the non-Gaussian noises was due to (nonlinear) interactions between fluctuators.

The stored charge in each logical node of a VLSI circuit decreases with decreased dimensions and decreased voltages in nanotechnology. Weak radiation can cause disturbance in the circuit signals, leading to increased soft-error failures. The fault/error-detection probability can serve as a measure of soft-error failure, which is an increased threat in the nanodomain logic node. In order to model the fault/error-detection probability, Rejimon and Bahnja (2005) presented a Bayesian network for error-sensitivity analysis in VLSI circuits. Their procedure provided a framework to obtain the joint pdfs of all variables in the network for calculating the error-detection probability.

As the technology node goes down to 90 nm and below, variability in device performance becomes a crucial problem in the design of ICs. In the past, die-to-die variability, which was well managed by the worst-case design technique, dominated the within-die variability. Onodera (2006) pointed out that the statistical nature of the variability has been changed such that the within-die variability is growing. This characteristic presents a challenge in circuit-design methodology. His paper provided measurable results of variability in 0.35-, 0.18-, and 0.13-μm processes and explained the trend of variability. It also showed that a circuit that was designed optimally under the assumption of deterministic delay is now most susceptible to random fluctuation. This result indicates the need of applying statistical thinking in the circuit-design methodology.

For solving process-variation problems in nanoscale-IC manufacturing, Li et al. (2005) proposed a novel projection-based extraction approach, PROBE, to efficiently create quadratic response surface models and capture both interdie and intradie variations with affordable computation cost. Instead of fitting a full-rank quadratic model, PROBE applied a projection operator and found an optimal

low-rank model by minimizing approximation errors, which were defined by the Frobenius norm. In PROBE, the modeling accuracy and parsimony can be tuned by increasing or decreasing the dimension of the projection space. Several examples from digital and analog circuit-modeling applications demonstrated that PROBE can generate accurate response surface models while achieving up to 12 times faster speed as compared with traditional process-variation simulation and modeling methods.

Stochastic Modeling

Due to the fact that a large proportion of variability in nano synthesis/growth processes are not well explained by known physical models, stochastic modeling techniques have served as an effective way in characterizing such processes.

Miranda and Jimenez (2004) proposed a stochastic logistic model based on a Wiener process for characterizing the breakdown dynamics of ultrathin gate oxides (at 2-nm level) and for understanding the effect of voltage stress on the leakage current. The model had two components: a deterministic term with a logistic function describing the mean failure behavior and a random term with a Wiener process representing uncertainties and noises. Throughout the model, the nano-material–degradation dynamics were captured using a small set of parameters.

To characterize degradation of leakage currents of 3-nm gate oxides, Hsieh and Jeng (2007) considered a nonhomogeneous Weibull compound Poisson model with accelerated stresses. The oxides were in square metal-oxide semiconductor (MOS) capacitors grown on p-Si. One hundred twenty capacitors were irradiated at three levels of ion density, and the leakage current versus gate voltage was measured before and after each irradiation step. Figure 6 shows the sketch of the leakage current of the gate oxides at three ion levels and two accelerated voltages. They provided maximum-likelihood estimates of model parameters and derived the breakdown-time distribu-
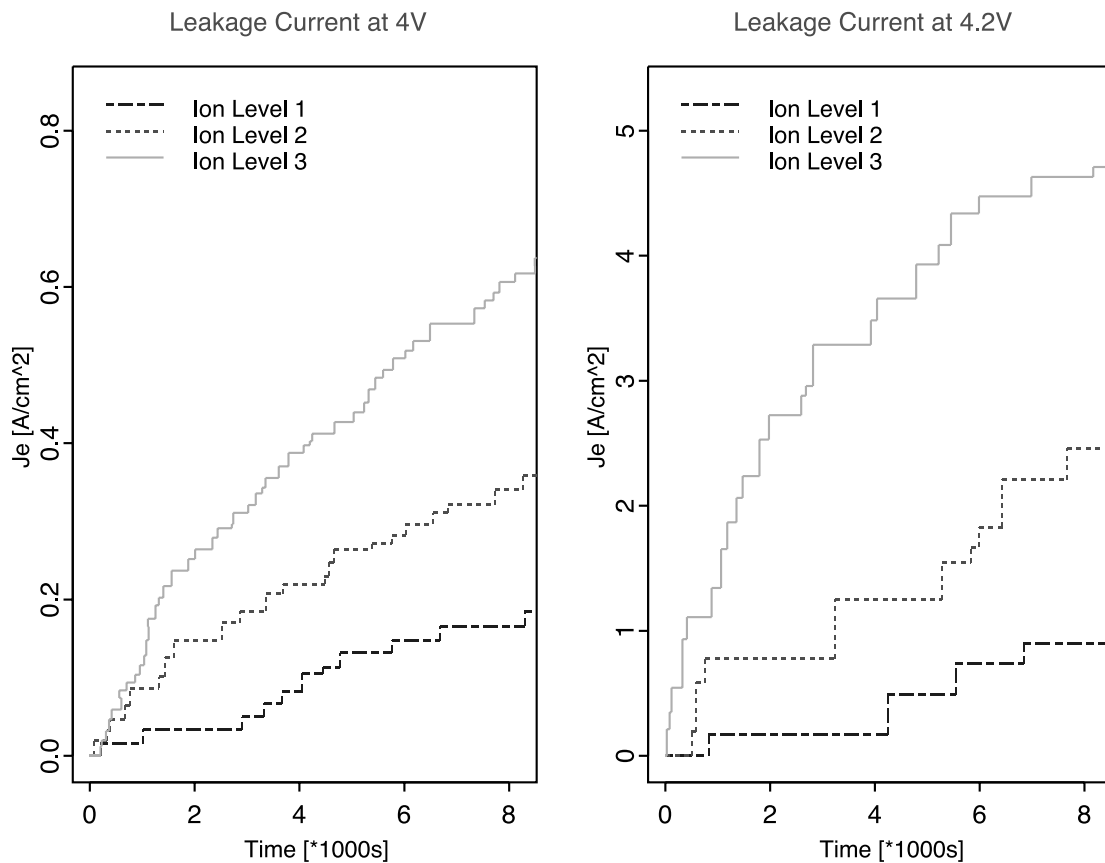

FIGURE 6. Leakage Currents of 3-nm Gate Oxides Under Accelerated Stresses (Voltages; from Hsieh and Jeng (2007)).

tion. To check the proposed models for the degradation measurements and the rate of breakdown-event occurrence, goodness-of-fit tests were considered. The estimated nanodevice reliabilities were calculated at lower stress conditions.

Time series of nanosystem measurements exhibit intermittency. At random times, the system switches from state *on* (or up) to state *off* (or down) and vice versa. Margolin and Barkai (2005) investigated the nonergodic properties of blinking nanocrystals modeled by a Levy-walk stochastic process. The process was characterized based on the sequence of on and off sojourn times. The times of the on–off events were mutually independent and were drawn at random from a pdf that followed a power-law distribution generated by a fractional Poisson process.

It has been widely recognized that novel nanoelectronic devices, such as carbon nanotubes and molecular switches, have high manufacturing defects due to the stochastic nature of the bottom-up physical and chemical self-assembly processes. Qi et al. (2005) reported that it was very challenging to manufacture nanocomputers with a device density of 10^{12} chips due to faulty components pervasive in the device. The authors studied the behavior of a NAND-multiplexing system with a Markov chain of distributions that could be unimodal or bimodal, depending on whether the probability of NAND gates was larger or smaller than a threshold value.

High-Frequency Signal and Spatial Data Analysis

Due to the use of advanced data-collection equipment, spatial data and/or high-frequency signals are collected in many nano applications. Because such data signals contain rich information about process status or product quality, effective (and efficient) statistical analysis methods are greatly important for extracting knowledge. The following paragraphs present a few examples.

Spatial statistical models have been used in modeling nanoscale structures. Chen and Lee (2004) used lattice points to represent atomic bonding units in a crystal. The structure of the unit together with the network of lattice points determined the crystal structure and the physical properties of the nanomaterial. In their experiments, polycrystalline solids consist of randomly distributed grains and grain boundaries. The size of grains was usually in the nano/microscale. Each grain was modeled as crystallized solid by micromorphic theory, while the grain

boundaries were modeled by classical continuum theory. Within each grain, the atomic motion led to the continuum lattice deformation from nanoscale to microscale. A multiscale spatial model was then used to characterize the material behavior of polycrystalline solids.

In order to understand the macroscopic properties of heterogeneous materials, modeling spatial correlations for microstructures is needed. Jefferson et al. (2005) analyzed microstructures of polymer nanocomposites and discovered that the material measurements did not exhibit randomness from a single homogeneous distribution. The traditional empirical model became inappropriate. A mixture of two distributions that defined the transition probabilities between two phases of the composites was introduced as a tool to examine the randomness and periodicity in the microstructure.

For manufacturing high-performance films with a thickness of only a few nanometers, nondestructive testing methods are required to ensure production quality. Schneider et al. (2002) developed a laser-acoustic technique based on surface acoustic waves for testing nanometer film's hard-coating quality. A nitrogen-pulse laser was used to generate a wide-band surface acoustic wave. When propagating through the nanofilm surface, the wave signals were recorded using a digitizing oscilloscope. Applying the Fourier transformation to the high-frequency laser-acoustic signals, the authors obtained empirical estimates of thickness and hardness of the films. Because the recent popular tool for modeling high-frequency signals is wavelets, it might be interesting to see the applicability of wavelets and the results they lead to in analyzing laser-acoustic signals. See papers in the subsection of process monitoring of functional data for examples.

Statistical Process Control (SPC) and Quality Control

As a traditionally popular technique in manufacturing, SPC is expected to serve the role of identifying assignable causes and thus reducing variations in nano-manufacturing processes. However, in this early stage of research in nanotechnology, nano-production systems have not matured yet. Hence, publications on SPC application to nano-technology research are scarce. This section highlights two important issues appearing in nano technology: functional data monitoring and feedback control. Although most of the feedback control studies deal with control-theory,

i.e., they are not statistically oriented, this review does dig out a few sensor-data–based control studies. In addition, several multiscale modeling and control publications illustrate the potential of future statistics research in the important nanotechnology development areas (see the description in the second generation of Figure 1). This review covers the latest progress on nano-SPC published in the following journals: *Computers & Chemical Engineering, Control Engineering Practice, IEEE Transactions on Plasma Science, IEEE Transactions on Semiconductor Manufacturing, Journal of Process Control, Journal of Quality Technology, Nanotechnology, Surface & Coatings Technology*, and *Wear*.

Process Monitoring of Functional Data

Functional data characterize quality or reliability performance of many manufacturing processes. They are very informative in process monitoring and controlling for nanomachining, ultrathin semiconductor fabrication, and several other manufacturing processes seen in the literature. In particular, wavelet analysis has been popular in modeling and monitoring functional data.

Ganesan et al. (2003) combined online detection with offline modeling strategies in the nanodevice wafer-polishing process. Through wavelet-based multiresolution analysis methods, the delamination defects were identified by analyzing the nonstationary sensor signals based on acoustic emission and coefficient of friction. Jiang and Blunt (2004) introduced a wavelet model to extract linear and curve-like features from complicated nonstationary surface-texture patterns in nanometer morphological structures. Through wavelet decompositions and reconstructions, Das et al. (2005) removed noises from the coefficient of friction signals in their chemical mechanical polishing of nanodevices and used a sequential probability ratio test to set up a control chart for monitoring process conditions.

Feedback Control

Sensor-Data–Based Control

Due to the ultrasmall size of objects and very fine manufacturing processes, using data collected from various sensors for developing precise automatic control rules is critical in nanotechnology studies. For instance, Ohshima (2003) pointed out that most traditional feedback controllers using existing sensors for micro and larger systems were not effective for dealing with nanomachines, where physical and chemical phenomena occur in very short time windows. The author considered *in-situ* feedback schemes to control physical–chemical reactions in their material processing courses. Klepper et al. (2005) developed an *in-situ* feedback–control tool to improve the reproducibility of a nanostructure deposition process. In order to reduce variability in the coating process, the investigators needed to find ways to measure process variations. Experiments were set up to test the assumption that plasma discharge was sensitive to the surface-roughness variation. The experiments resulted in the identification of the correlation between hydrogen atomic emission and formation of metal-carbide coatings. Through the control of the emission, variability in the nanostructure composition and the wear performance of the coating was controlled by a closed-loop feedback algorithm.

Lin et al. (2003) studied precise positioning issues in a nanoscale drive system. To guarantee the desired precision, the authors used an integral type of controller in the positioning. The study demonstrated the possibility of achieving a very precise positioning at the resolution level of sensor measurements. Lantz et al. (2005) introduced a novel micromachined silicon-displacement sensor with displacement resolution of less than 1 nm for providing accurate positioning information. This new technique can detect the displacement by measuring the difference between the resistances of the two sensors.

Multiscale Modeling and Control

A few factors, such as the following, motivate the current active research on control of multiscale processes (see the preface of the special issue on control of multiscale and distributed process systems in 2005, volume 29, *Computers and Chemical Engineering* for details).

(a) key technological needs in semiconductor manufacturing, microtechnology, nano technology, and biotechnology; and

(b) recent developments in actuator and sensor technology that make control of material microstructures, spatial profiles, and product-size distributions feasible and practical.

Using the thin-film growth process as an example, Christofides and Armaou (2006) provided a review of recently developed methods for controlling multiscale processes. The typical thin-film growth process has been widely used in microelectronic devices, nanomaterials, and nanocomposites (see, e.g., Ng et al. (2003), Sneh et al. (2002), Mae and Honda (2000)).

To achieve better control of multiscale processes, process modeling and prediction by using physical, statistical, or simulation methods to characterize complicated process outputs becomes a critical issue. The following provide a few examples.

Precise control of film properties in nanoscale semiconductor manufacturing requires models that predict how the film state (in the microscopic scale) is affected by changes in controllable parameters (in the macroscopic scale). Lou and Christofides (2003) developed a multiscale model that involves coupled partial differential equations (PDEs) for modeling the gas phrase of the material deposition process. Subsequently, the authors used a kinetic Monte-Carlo (kMC) simulator to model the atom adsorption, desorption, and surface-migration processes for shaping thin-film microstructures. There are strong interactions between the macro- and microscale phenomena. For example, the concentration of the precursor in the inlet gas governs the rate of adsorption of atoms on the surface, which, in turn, influences the surface roughness. On the other hand, the density of the atoms on the surface affects the rate of desorption of atoms from surface to the gas phrase, which, in turn, influences the gas concentration of the precursor.

Lou and Christofides (2003) constructed an efficient estimator for the surface roughness at the timescale comparable to the real-time evolution of the process using discrete on-line measurements. Then, the estimated roughness was fed into a proportional-integral (PI) controller. Application of the estimator/controller to the multiscale process model demonstrated successful regulation of the surface roughness at the desired value.

In nanoscale semiconductor manufacturing, the kMC simulation methods have been used for

1. predicting microscopic properties of thin films (e.g., surface roughness);

2. studying the dynamics of complex material deposition processes including multiple chemical species with both short-range and long-range interactions; and

3. performing predictive control design to control final surface roughness.

kMC is not available in closed form, thus making it difficult for use in system-level analysis and design and implementation of model-based feedback-control systems.

Instead of using the kMC approach in develop-

ing feedback-control schemes, for many deposition and sputtering processes, a system of stochastic linear/nonlinear PDEs is derivable based on microscopic rules corresponding to the so-called master equation, which describes the stochastic nature of the thin-film growth process (Van Kampen (1992)). Lou and Christofides (2006) proposed using nonlinear stochastic ordinary differential equations (ODEs) (an approximation of the stochastic PDEs) for developing computationally efficient feedback controllers. Their objective was to control the expected roughness of the surface. The solution strategy is to control the covariance of the thin-film growth states in the nonlinear stochastic PDE system for various spatial locations. Based on various simulation runs, it is clear that the proposed nonlinear feedback-control method can reduce the surface roughness to the desired level, while also effectively reducing the variance of the final surface roughness.

Different control strategies have been used to control multiscale processes. Both proportional-integral (PI) controllers (Lou and Christofides (2003), Lou and Christofides (2006), Gallivan (2005)) and model-based predictive controllers (Ni and Christofides (2005a, b), Christofides and Armaou (2006)) have been implemented in practice to control thin-film growth surface roughness and other parameters of interest. Figure 7 shows a general framework of predictive controllers. The difference between the real and predicted output are compared; such information is then fed into the optimizer to generate an optimal set point for the next step. Multiscale models
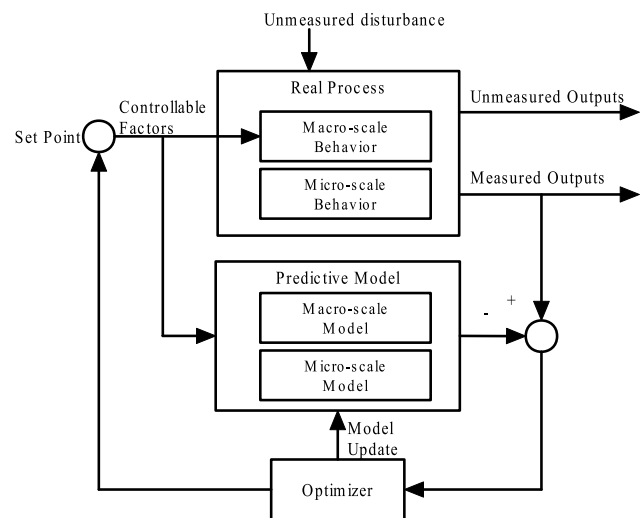

FIGURE 7. A Framework of Predictive Controllers with Multiscale Models.

can be employed in building the predictive model. In nano applications, a multiscale model can be used to characterize both macroscale and microscale behavior of a process. Usually, manipulated variables are applied to only the macroscale behavior of a process. Formulating an objective function is a key task in deriving the optimal set point. Most objectives in the multiscale control publications focused on the upper scale process performance. See Fenner et al. (2005) for a study of incorporating objectives from multiple process stages in deriving an automatic-control policy. The following gives another example.

The use of PI or predictive controllers has successfully improved process stability and product quality. However, in controlling a nanopositioner device, Salapaka et al. (2002) noted that the conventional PI controller does not meet the requirements for positioning. Instead, the authors considered an H-infinity controller to incorporate both performance and robustness in the objective function and found the optimum based on the H-infinity norm. As the objective is defined on multiple scales, the H-infinity norm is calculated as the supremum of the objective function on all scales. The optimization method based on H-infinity is a feasible way to solve multiscale problems and has substantially improved positioning speed and precision in the research.

Conclusion and Future Research

This article has reviewed applications of statistical techniques in nano technology. As the physical and chemical properties of nanoscale materials differ fundamentally from the properties of individual atoms and molecules or bulk matter, various challenges have occurred in designing, analyzing, and manufacturing nanodevices. Therefore, both conventional and new statistical techniques are expected to play important roles in nanotechnology research.

This paper first investigates the experimental design issues of nanodevice fabrication processes. Diverse application examples include fractional factorial designs, response surface designs, and robust parameter designs. For future nanotechnology research to make efficient use of these methods, we feel that the following issues are worthy of consideration:

(a) *Choice of experimental-design methods.* In some applications, simply full factorial or fractional factorial designs may be sufficient for model building and process understanding. However, as shown in several examples, nanofabrication processes can be very sensitive to small changes in controllable and noise factors. Moreover, the process outcomes may not be continuous random variables. In this case, irregular designs or new experimental design methods might be considered. Finally, special experimental constraints due to physical limitations may limit the use of certain designs.

(b) *Analysis of experimental data.* Due to the use of advanced measurement tools, spatial data, high-frequency signals, and/or qualitative measures are frequently observed in nano research. Combined with the multistage nature of the nanodevice–fabrication processes, statistical analysis of such complicated and/or large size data with many explanatory and noise factors provides many challenges. New algorithms, such as those developed by Jeong et al. (2006), Yuan and Kuo (2006), and Wang and Tsung (2007), might be useful for dealing with these new data types. Variable selection tools, such as Yuan et al. (2007), could be effective for dealing with large size factors constrained with experimental design structures. However, more research is needed in this field, especially for multistage, multiscale, and multilevel processes.

(c) *Use of computer simulations.* When the physical experimentation is costly and time consuming, the use of computers for process and device simulations is expected to become more prevalent in nano research. Sometimes, processes can be very complicated, leading to lengthy computer experiments. Thus, the design and analysis of computer experiments, as well as the integration between computer and physical experiments, are topics that deserve more future research efforts.

(d) *Interaction between statisticians and experts in material science, physics, and other disciplines.* Nanotechnology is a multidisciplinary subject. When statisticians are more involved in understanding the issues in nanoresearch, more effective procedures can be developed to deal with challenging application problems. Statisticians should be important players in novel scientific discoveries.

Quality issues have been emphasized in early production stages of nanodevices. The challenges confronted when applying SPC and APC techniques in nano applications are confounded with the new data types discussed above. Moreover, when online sensor information is available, it is interesting to see the of-

fline robust parameter designs extended (e.g., Joseph (2003)) to take advantage of the new information useful for process adjustments. See Edgar et al. (2000) and Del Castillo (2006) for a review in the topics of automatic control and statistical process adjustment.

Research on reliability is also critical to nanodevices and their fabrication processes. The intrinsic mechanism of failures for nanoproducts and the modeling of their lifetimes are areas of future research interest. Readers may refer to Jeng et al. (2007) for a detailed review in nanoreliability studies and see other papers in the same issue of the journal for specific topics. A different, but related, direction of new research is the "reliability" of measurement and positioning systems in nanomanufacturing. Because nanodevices are so small and the nanomanufacturing process requires speedy data collection, how can one know if the measurements are accurate and, more important, if the nanosubsystems are placed in the correct location within a very small-sized nanosystem? See Xia et al. (2007) and Qian and Wu (2007) for examples of new statistical methods developed for integrating information with different accuracy for statistical inference.

With the fast development of nanotechnology and the introduction of mass production of nanodevices, statistics is expected to play an increasingly important role in both academic and industrial fields. This review summarizes successful applications of statistical methods in nanotechnology seen in the literature, along with a few interesting topics for future research.

Acknowledgments

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