Challenges and Needs for Automating Nano Image Processing for Material Characterization

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ABSTRACT

Image processing techniques are needed to extract critical information pertinent to nano material characterization but the current processing methods are slow, expensive and labor intensive. There is a strong need to develop fast and reliable methods, enabling process control compatible automated processing of nano images. The authors believe specialized techniques are needed to address the challenges, and will discuss the recent development of nano image processing methods as well as the near- and medium-terms needs in the area of nano metrology and imaging. The authors will share their broad perspectives on this research direction.

Keywords: Electron microscopy, nano metrology, quality and process control, scalable nanomanufacturing.

1. INTRODUCTION

National Nanotechnology Initiative (NNI)'s signature initiative [1] identified the major technical roadblocks toward the goal of scaling up nanomanufacturing (SNM). The second roadblock in the NNI signature initiative is the nanomanufacturing measurement technologies that can satisfy the needs for process and quality control of SNM processes. In this paper, we intend to discuss the research challenge and needs in the context of automated nano image processing for the purpose of material characterization, a task we believe of essential importance in addressing NNI's Signature Initiative**.**

The NNI Signature Initiative states: "*Existing [measurement] methods are time-consuming, expensive, and require high-tech infrastructure and high skill levels to perform*." Our own experience confirms what is said in this statement. Working with various nanomanufacturing processes, we realize that measurements revealing the structure and function of nano subjects are most likely in an image format. Consequently, image processing techniques are inevitably needed to extract critical information pertinent to a subject's formation as well as interaction with the host materials. These images (referred to as nano images hereafter) are of large data size and contain complicated features. The current nano image processing methods are indeed slow and laborious; it sometimes takes hours to complete the processing, while for in-process control purposes, the processing time needs to be within the range of a few minutes. NNI's statement calls for innovation in developing both hardware metrology devices and software measurement processing methods that meet the fast, reliable, process-control-compatible requirement.

The international standard on nanotechnology leaves it for the users to identify an appropriate data analysis algorithm or to seek a third party software solution. When an incapable image processing tool is used, this approach leaves a gap in the loop of nano metrology on imaging data. The high resolution enabled by the physical instruments may not be preserved.

We also believe that nano images have their uniqueness. Features valid, and techniques favored, in the general image processing or bio-imaging field may no longer be effective in nano imaging. Specialized image processing techniques are needed to address the challenges. Towards that goal, we discuss in this paper our recent development of image processing methods specifically tailored for the nano image data, as well as the near- and medium-terms

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needs in the area of nano metrology and imaging.

The paper unfolds as following. Section 2 describes the state of the art practice in processing nano images. Section 3 presents a summary of our recent development of specially tailored methods for processing nano images. Section 4 discusses the research needs in the near future. Section 5 summarizes the paper.

2. STATE OF THE ART PRACTICE

Nano images contain the morphological information (size and shape) of nano subjects as well as the information of their locations; the latter can be used to discern the uniformity of dispersion or distribution of nano subjects in a host material. The state of the art practice can be summarized in the following three aspects:

(a) **General image processing methods and bio-imaging methods**. We come across a good number of existing methods, including watershed methods [2-5] (in the mathematical morphology branch [6]), active contour [7-10], graph cut [11,12], sliding band filter [13,14], iterative voting [15], and a sophisticated multi-scale decomposition approach [16] that was developed rather recently. Many variants and recently improved versions of these methods are also investigated [17-20]. A summary of these methods and their performance comparison based on a set of gold nanoparticle micrographs can be found in [21].

(b) **Nano imaging in the fields of material sciences and manufacturing**. There is a limited amount of literature [22-25] about automated analysis of nano images in the fields of material sciences and manufacturing. Existing methods in the fields of material and manufacturing are less sophisticated than those developed in the general image processing or bio-imaging field. They often make strong simplifications, invoke heuristics, or simply adopt techniques from the image processing field as is. Here we articulate "automated" because *manual analysis* of material images for characterization was conducted immediately after imaging equipment was introduced to material sciences.

(c) **Software and tools used in practice**. There exist a few software tools, some of which come with the characterization instruments, such as a transmission electron microscope or scanning electron microscope. When our team informally surveyed several nano-research groups in and outside the U.S, including those working with us, we find that one popular tool in use is ImageJ [26]. ImageJ is a freeware provided by the National Institute of Health for cell morphology analysis. Given the apparent similarity between cell and nano images, it is not surprising that people went to the bio-imaging field to look for a tool. ImageJ is a good representation of the tools used in practice because other commercial software tools we came across do not really exhibit any greater capability than that of ImageJ. ImageJ does have a number of plug-ins, which are built based on the methods reviewed above in (a).

The current image processing methods are ineffective for scalable production purpose, due to its low recognition rate of nano subjects. One principal challenge is due to the existence of overlaps between nano subjects observed in nano images, meaning that a cluster of nano subjects often aggregates together, forming an agglomerate. This challenge is made even more difficult by a number of other factors, such as the sheer number of nano subjects in a nano image (in hundreds or thousands), a great variety of geometric shapes that can be taken by nano subjects (round, triangle, rectangle, or rod), high noise and low contrast if nano subjects are embedded in a host substrate of similar atomic masses, and the lack of underlying truth to guide a learning algorithm.

When the existing methods are applied to typical nano image, one finds that over-segmentation and under-segmentation occur frequently. For instance, as reported in [27, Fig. 12], when it is used on a nano image in which nanoparticles are moderately overlapping, ImageJ only identifies 28% of the particles correctly, or 110 out of the 396 total. This low rate of recognition will have to be compensated by laborious manual operation to isolate nano subjects, because without isolating individual subjects, neither morphology analysis nor dispersion quantification (i.e., assess how uniformly nano subjects are distributed in the host material) can be accurately performed. It is evident that the lack of automation in processing the nano images creates a bottleneck hampering scale-up towards the creation of an industry of tomorrow.

3. RECENT DEVELOPMENTS

Handling overlapped nano subjects entails three major technical undertakings (see Figure 1): (a) *image segmentation*, aiming at separating individual subjects from a nano-subject agglomerate; (b) *contour inference*, recovering the missing parts of the separated nano subjects; (c) *shape classification*, classifying the nano subjects by shape and compute its morphology related metrics (such as size or aspect ratio). Fulfilling the research objectives needs advancement not only in image processing [28] but more so in the intersection of image processing and statistical shape analysis [29]. In the past, however, research conducted in the two fields has been done by and large sequentially; image processing first and then shape analysis. This may have presented a fundamental obstacle causing the lack of reliable methods in processing the large number of overlapped subjects of various shapes present in nano images.

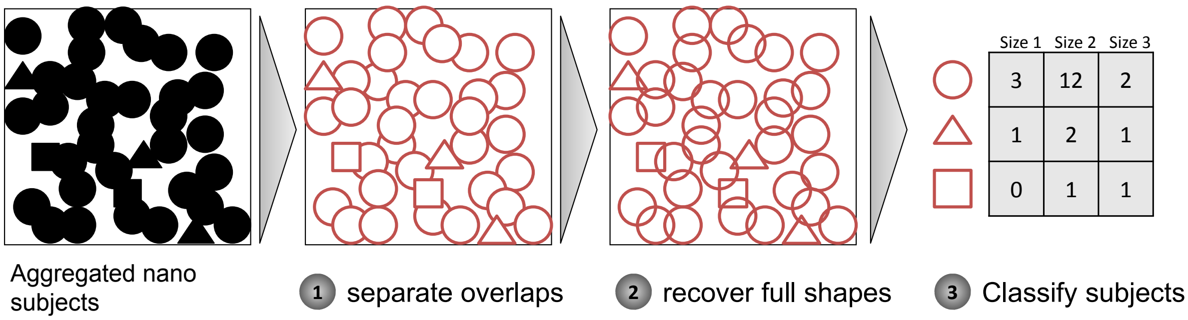

Figure 1. Three major technical components in isolating nano subjects for analysis and characterization.

We have investigated and developed several approaches of different flavors [21, 27, 30, 31]. We started off with a three-stage, divide-and-conquer strategy [27] and gradually evolved to more sophisticated, integrated approaches [21, 30]. Our most recent work handles as well the high-level noise existing in some of the nano images [31].

Three-stage, divide-and-conquer approach
(IIE Transactions - Quality & Reliability [27])

- Particle separation via a convexity analysis;
- Recover missing contour using functional principal component analysis (PCA) [32]
- Shape classification using the k-nearest neighbor method [33].

Bayesian shape analysis approach
(Annals of Applied Statistics [30])

- Nano subjects modeled after a set of candidate shapes in a dictionary;
- Shape evolution represented by a reverse jump process [35];
- Bayesian solution finds the optimal match between a shape template and image observation.

Two-stage, simultaneous learning approach
(IEEE T-Patten Analysis & Machine Intelligence [21])

- Segmentation via a robust, revised watershed variant;
- Joint learning of missing contour and shape class;
- A B-spline contour regulated through a Gaussian mixture model and solved through an ECM algorithm [34].

Robust fusion of complementary image infomation
(IEEE T-Image Processing [31])

- Both image intensity and gradient information are utilized;
- Consensus detection reinforces the trust of a correct identification.
- Conflict detection are resolved through a binary integer programming (IP) formulation [36] and solution.

Figure 2. Summary of different approaches investigated in the team's preliminary study.

Figure 2 summarizes the features of the approaches in our research development. Our current methods made a remarkable improvement. For instance, in the example mentioned in Section 2, of which ImageJ identified correctly only 28% of the 396 nanoparticles, one of our approaches, reported in [27], can identify over 90% of them correctly. From the summary in Figure 2, one can also observe that a wide variety of image processing, machine learning and statistical modeling techniques were used and tailored for the nano imaging problems. It is our belief that in order to effectively address the challenging problem of automated nano image processing, the latest developments and tools in all relevant areas must be fully made use of, so that the unprecedented capability called for could possibly materialize and the technical barriers be overcome.

4. RESEARCH NEEDS

Our recent developments have made remarkable inroad towards making nano imaging methods compatible with scaled-up production, especially in terms of addressing the uniqueness of nano images and boosting the identification rate of entangled nano subjects. To make the nano imaging techniques ready for production scale, we believe that several additional challenges have to be addressed:

- **Enhancement of computational efficiency**. The computational efficiency of handling nano images with over 1,000 nano subjects still needs a boost. The current methods take anywhere from tens of minutes to close to an hour to process a nano image of such scale. For a method suitable for in-process quality control purpose, the processing time needs to be at the level of a few minutes.
- **Assessment of metrology characteristics**. Nano imaging is one critical element of the whole nano metrology process. Outcomes of nano imaging (the measurement processing methods) need to be reflected in the final metrology assessment, such as characterization of bias, repeatability and reproducibility, so that one can quantify how much a quality engineer can trust the output from the nano metrology equipment.
- **Sequential sampling based on multi-resolution measurements**. One nano image of 1,024-by-1,024 pixels under nanometer resolutions covers a very small region of bulk materials, e.g., a 100 nm-by-100 nm view field. To ensure the quality of the whole material, a single nano image is insufficient. But surveying the whole bulk in the same nanometer resolution is not practical, as it would take forever to analyze even a single piece of product, hardly something quality engineers could use for continuous production. We hypothesize that a better approach is to trade off measurement speed and accuracy by studying nano images taken at different resolutions (such as at 10, 100, and 1,000 nm). Understandably, the measurements taken at the low resolution are faster but less specific, while the high-resolution measurements are slower but revealing. An efficient approach could be to let the low-resolution measurements to guide a sequential sampling over the bulk material and triggers high-resolution measurements only when necessary.
- **From 2-d to 3-d**. Up to date, the vast majority of nano images used in material characterization are two-dimensional projections of nano subjects that are embedded in a three-dimensional space. The inverse problem is how we can infer about the nano subject's morphology and distribution in the original three dimensional space. Electron microscopic tomography [37] is now available, and reconstruction can be done based on the Fast Fourier Transform (FFT) and Inverse FFT [38]. But the key difference here is that those reconstructions are still pixels based, meaning that to interpret features, human experts are needed to look at the reconstructed images (similar to what doctors do with medical computerized tomography). Two problems are of interest to explore in the near term: one is how much we can say about 3-d characterization based on 2-d images taken at a single projection angle; and the other is about the same question but based on a sequence of 2-d images obtained at different projection angles.
- **Imaging moving and morphing nano subjects**: In recent years *in situ* techniques such as x-ray scattering, TEM, SEM, and laser-based optical imaging have been introduced for monitoring nanomanufacturing processes [39-45], such as sol gel processes for nanoparticle and self-assembled cluster synthesis, as well as chemical vapor deposition processes for synthesizing nanotubes, wires and films. These techniques provide an opportunity to monitor, at atomic resolutions, the motion and transformations that the individual nano subjects undergo during a nano synthesis process (i.e., estimate process state). Ability to track the motion and transformation of individual nano subjects at high specificity can accelerate the discovery of causal pathways that lead to various nanostructures [46]. It can also provide a precise assessment of the effects of various process parameters, compared to the current, population-based statistical descriptions of the transformations (e.g., average change in particle size). Towards this end, the nano subjects need to be tracked not just within a synthesis step but also over multiple stages of a nanomanufacturing process (e.g., catalyst film deposition,

stabilization through the nanostructure growth stage). Two challenges that arise in this context are — how to associate nano subjects over multiple image snapshots? What time-resolutions are needed for different imaging techniques-nanostructure combinations to achieve a specified level of confidence with identifying individual nano subjects? Intuitively, these challenges are akin to those encountered in identifying persons of interest based on multiple facial images, albeit over a much short time scales and more rapid transformations.

5. SUMMARY

In this paper, we present our perspectives concerning nano imaging and its future research for the purpose of satisfying the needs in scalable nanomanufacturing. This line of research is to address one major roadblock identified by NNI's signature initiative and meant to benefit a broad array of scalable nanomanufacturing processes, as opposed to a specific process. The proposed nano imaging capability, if materialized, could enable process and quality control capabilities in nanomanufacturing processes, leading to reduction in waste of energy and materials and improvement in efficiency. A competent quality control is essential to reducing a manufacturing system's negative footprint on environment and to enhancing its market competitiveness. Our final note is to point out that our discussion here focuses on the software/algorithm side, namely, the development of efficient measurement data processing methods. We want to stress that to fulfill the process control objective, innovations in both measurement technology (hardware) and data processing capability (algorithms) are pressingly needed.

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