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Accurate segmentation of dense nanoparticles by partially discrete electron tomography

T. Roelandts [a,*], K.J. Batenburg [a,b], E. Biermans [c], C. Kübel [d], S. Bals [c], J. Sijbers [a]

[a] IBBT-Vision Lab University of Antwerp, Universiteitsplein 1, 2610 Wilrijk, Belgium
[b] Centrum Wiskunde & Informatica, Science Park 123, 1098 XG Amsterdam, The Netherlands
[c] EMAT, University of Antwerp, Groenenborgerlaan 171, 2020 Antwerp, Belgium
[d] Institute of Nanotechnology, Karlsruhe Institute of Technology, Hermann-von-Helmholtz-Platz 1, 76344 Eggenstein-Leopoldshafen, Germany

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ABSTRACT

Accurate segmentation of nanoparticles within various matrix materials is a difficult problem in electron tomography. Due to artifacts related to image series acquisition and reconstruction, global thresholding of reconstructions computed by established algorithms, such as weighted backprojection or SIRT, may result in unreliable and subjective segmentations. In this paper, we introduce the *Partially Discrete Algebraic Reconstruction Technique* (PDART) for computing accurate segmentations of dense nanoparticles of constant composition. The particles are segmented directly by the reconstruction algorithm, while the surrounding regions are reconstructed using continuously varying gray levels. As no properties are assumed for the other compositions of the sample, the technique can be applied to any sample where dense nanoparticles must be segmented, regardless of the surrounding compositions. For both experimental and simulated data, it is shown that PDART yields significantly more accurate segmentations than those obtained by optimal global thresholding of the SIRT reconstruction.

1. Introduction

Electron tomography deals with the reconstruction of a three-dimensional (3D) representation of a microscopy sample from a tilt series of two-dimensional (2D) images. This technique has been applied successfully in materials science since the late 1980s [1]. Several imaging modes have been used for acquiring the projection images, in particular, bright-field TEM [2,3], annular dark field TEM [4], high-angle annular dark-field scanning TEM (HAADF STEM) [5–9], and energy-filtered TEM (EFTEM) [10–13].

Quantitative interpretation of the reconstructed 3D volume is often hampered by the presence of *artifacts*: structured distortions that do not correspond with the actual sample. In particular, limits on the number of projection images imposed by sample contamination or beam damage give rise to such artifacts. Furthermore, the limited spacing for specimen holders in between the pole pieces of the objective lens often restricts the range of tilt angles to about $\pm 70^{\circ}$, leading to a missing wedge in the collected data. As a consequence, features perpendicular to the electron beam are better resolved than features parallel to the beam, resulting in anisotropic resolution and distortions of the structure.

For many imaging tasks in materials science, the goal is to obtain an accurate *segmentation* of particular structures (i.e., particles, pores, tubules, etc.). Of particular importance is the problem of segmenting nanoparticles within various matrix materials [5–7,14]. Due to artifacts related to image acquisition and reconstruction, segmenting these structures from gray level volumes computed by established algorithms, such as weighted backprojection (WBP) or SIRT [15], may result in unreliable and subjective segmentations. In practice, reconstructions are often segmented using a global threshold. Since the threshold is estimated visually, this approach is highly subjective. Moreover, it does not account for the effect that the intensity of the features in the reconstruction strongly depends on their size [16]. Fully manual segmentation may avoid this effect, but remains a time consuming and subjective approach.

Recently, discrete tomography algorithms have demonstrated the ability to overcome some of these limitations by exploiting prior knowledge. Discrete tomography is based on the assumption that the sample consists of only a few different compositions. Two rather different variants of discrete tomography have been applied to electron tomography. The first variant was recently applied to the reconstruction of crystalline nanoparticles at atomic resolution [17,18]. For this variant, it is assumed that the crystal contains only a few atomic species, and that the atoms lie on a regular grid. Together, these assumptions allow to create a reconstruction from as few as two or three projections. For the second variant, which can be applied at lower resolutions, it is

* Corresponding author. Tel.: +32 32652840; fax: +32 32652245.
E-mail address: tom.roelandts@ua.ac.be (T. Roelandts).

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only assumed that the sample consists of a few different compositions, each corresponding to a particular gray level in the reconstructed image. The discrete tomography algorithms that appear in this paper are of the second variant.

Major advantages of discrete tomography algorithms are that they require fewer projection images compared to alternative methods such as SIRT, and that missing wedge artifacts are strongly reduced [19]. Moreover, as the final result of the reconstruction process is a segmented image, a separate segmentation step is no longer required. The *Discrete Algebraic Reconstruction Technique* (DART) for discrete tomography has been successfully applied to a broad range of material samples [20–23]. The main restriction for using discrete tomography is that the entire sample must satisfy the discreteness requirement. If the sample contains a mixture of compositions, the results of discrete tomography cannot be relied upon, as the key assumptions are violated.

In this paper, we introduce the *Partially Discrete Algebraic Reconstruction Technique* (PDART) for computing accurate segmentations of dense nanoparticles of constant composition, regardless of the compositions in the remaining part of the sample. Embedded nanoparticles such as catalyst particles are often dense structures compared to their surroundings (e.g., porous materials), resulting in a high gray level in the reconstructed image. PDART is based on the assumption that the densest composition occurs in homogeneous regions that have a constant gray level. These dense regions are segmented discretely, while the surrounding regions are reconstructed using continuously varying gray levels. If the assumption of a homogeneous densest composition holds, the imaging mode that is used to record the tilt series – HAADF STEM for both samples in this paper – is not a restriction on the applicability of PDART, as long as the selected imaging mode is compatible with tomography. PDART imposes no restrictions on the nature of the sample (except that the densest composition must be homogeneous), which means that the application of the algorithm is not restricted to any specific type of samples.

This paper is structured as follows. In Section 2, the problem of segmenting dense particles is introduced, and the PDART algorithm is defined. Section 2 also introduces the figure of merit that is used for quantitative evaluation of the results. It concludes by describing how the parameters of the algorithm can be optimized automatically. In Section 3, the capabilities of PDART are assessed using two different experimental datasets and a number of simulation experiments. The results are discussed in Section 4 and conclusions are drawn in Section 5.

2. Algorithm

Before describing the PDART algorithm, we start by giving an example of its applicability. Fig. 1 illustrates the problem of nanoparticle segmentation. Fig. 1a shows a *phantom* (i.e., a simulated image), representing a microscopy sample that contains nanoparticles of only a few pixels each, embedded in a cylinder of varying composition. From this phantom, a synthetic dataset was created by calculating 28 evenly spaced projections in the range of $\pm 70^{\circ}$. Fig. 1 also shows WBP (Fig. 1b), SIRT (Fig. 1c), DART (Fig. 1d), and PDART (Fig. 1e) reconstructions of this dataset.

The gray level reconstructions computed by WBP and SIRT have limited visual quality, as a result of the small number of projection angles and their limited angular range. When thresholding these images to determine the size and shape of the particles, it is not clear how the threshold should be chosen in an optimal way. The DART reconstruction, shown in Fig. 1d, is already segmented, yet the segmentation is not accurate at all when compared to the original phantom. The varying composition of the disk surrounding the nanoparticles violates the key discreteness assumption imposed by the DART algorithm. The PDART reconstruction, shown in Fig. 1e, seems much more accurate than the other reconstructions.

2.1. Algorithm description

The PDART algorithm has been designed to allow for accurate particle segmentation in cases where neither continuous methods nor fully discrete tomography leads to good results. The algorithm is based on the assumption that the particles have a constant composition, and that this composition represents the highest gray level in the reconstructed volume. PDART combines an iterative reconstruction algorithm, such as SIRT, with intermediate segmentation steps. Once pixels have been identified as "particle", they are directly segmented (i.e., their value is set to the constant gray level for the particles) and kept fixed at this value in subsequent SIRT iterations. Note that, throughout this paper, we use the additive variant of SIRT, as described in [24].

Fig. 2 shows a flowchart of the PDART algorithm. Besides having the projection data as input, the algorithm has two parameters: a threshold τ and a gray level $\rho > \tau$, which corresponds to the gray level of the particles. Optimal values for both parameters can be determined automatically, as is outlined in Section 2.3.

Initially, the set F of *fixed pixels* is empty. In an iterative loop, the algorithm starts by performing one or more SIRT iterations on the entire image volume. Whenever one or more pixels are assigned a higher gray level than the threshold τ, it is decided that these pixels belong to a particle. Such pixels are added to F: their gray level is set to ρ and is kept fixed at this value during all subsequent SIRT iterations. In this way, the set F gradually expands as pixels are added, until some termination condition is satisfied. Typically, one aims for terminating the algorithm when no new pixels have been added to F for a sufficiently large number of iterations.

In its original form, the SIRT algorithm computes a weighted least square solution of the system $\boldsymbol{Wx} = \boldsymbol{p}$, where $\boldsymbol{x}$ denotes the unknown image, $\boldsymbol{p}$ denotes the projection data, and $\boldsymbol{W}$ denotes the

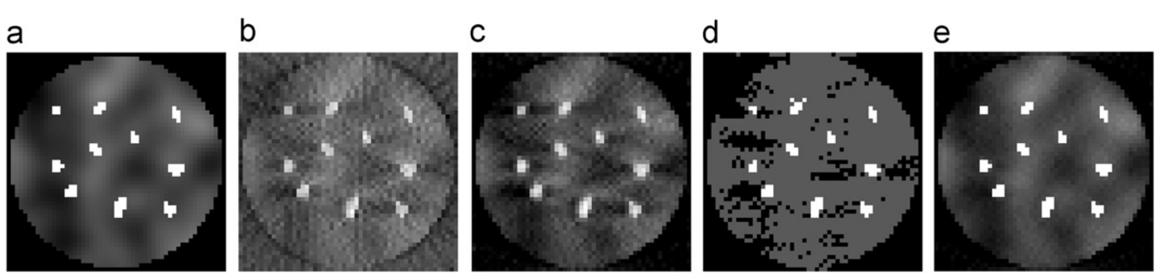

Fig. 1. A simulation phantom and several reconstructions. The phantom represents a cylindrical sample that contains nanoparticles of only a few pixels each, embedded in a material of varying composition. (a) Phantom, (b) WBP, (c) SIRT, (d) DART and (e) PDART.