DETERMINATION OF THICKNESS REFINEMENT USING STEM DETECTOR SEGMENTS

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Abstract

Quantitative STEM imaging together with Monte Carlo simulations of electron scattering in solids can bring interesting results about properties of many thin samples. It is possible to determine thickness of a sample, to calculate mass of particles and measure mass per length/area. Appropriate calibration is one of the crucial parts of the method. Even a small error or inaccuracy in detector response to electron beam - either blanked or full - brings significant error into thickness determination. This problem can be overcome by parallel STEM imaging in more segments of the detector. Comparing more segments gives a possibility to use a signal from different segments for different thicknesses of a sample. Accuracy of individual parts of the detector depends on the captured signal quantity. It is desirable to use such a STEM detector segment that provides the greatest signal change to a unit of thickness. To demonstrate the usage, we used a sample of Latex nanospheres placed on thin carbon lacey film; diameter of the nanospheres was around 600 nm in order to compare the results from different detector segments. Thanks to the known thickness of the sample (calculated from its geometrical shape), it is possible to estimate the optimal acquisition settings and post processing steps with the known and the true state of the sample.

Keywords: Quantitative STEM, thickness determination, detector segments, Monte Carlo simulation

1. INTRODUCTION

The field of electron microscopy offers many types of imaging techniques. In general, it is possible to divide imaging techniques into two groups by dimensionality of provided information. The first group gives 3D information about the investigated sample. It can be focused ion beam milling (FIB-SEM), serial block face imaging (SBF-SEM) or volume scope (deconvolution of images captured at different acceleration voltages) in the case of scanning electron microscopes (SEM). In the case of transmission electron microscopy (TEM) it is possible to use tomography or single particle analysis workflows. The second group is imaging in 2D either conventional imaging in SEM using of variate detectors and signals or planar projection of a sample in TEM and STEM. Quantitative scanning transmission electron microscopy (STEM) imaging brings a way how to get more quantitative information about the thickness, such as a thickness of the sample at each pixel from a single image (other available information may be a mass per length of filamentous structures, mass per area of sheets, or mass of particles). The main assumptions for this technique are 1) known geometry of the detection system that usually contains the geometry and position of the detector, the sample holder and the pole piece of the final demagnifying lens 2) density and composition of investigated samples. As clear from principle of STEM imaging, the samples are mounted on TEM grids. It depends on the type of the sample if the supporting film is used. Provided that it is necessary e.g. for imaging of individual nanoparticles, it is desirable to use a film as thin as possible with sufficient endurance to the electron beam. Correlative quantitative imaging in SEM and STEM is described in [1].

2. MATERIAL AND METHODS

Method based on workflow described by Volkenandt et al in [2] was used in this study. The main assumption is **a *linear response of a detector to intensity of incident electron beam***. It is necessary to calibrate the

detector system response on incident electron beam over the whole range of used probe currents. We used commercially available STEM III detector mounted on Magellan 400L SEM (both FEI, Czech Republic). Images were taken by the original internal scanning unit of the microscope with a dwell time of 5 µs and image size of 1024 × 882 pixels. Five amplifier lines were used for simultaneous acquisition of four independent signals: bright field (BF) - one channel, dark field 1 (DF1) - one channel, dark field 3 (DF3) - one channel, and high angle annular dark field (HAADF) - two channels. Dark field 2 and dark field 4 were not used in this study. Other SEM settings are as follows: acceleration voltage 30 kV, probe current 25 pA, working distance (WD) 4 mm, field free mode, pixel size approximately 1 nm. Quantitative imaging in immerse high resolution mode (described in [3]) was not used for its uncertainty and difficulty.

For Monte Carlo (MC) simulation of signals captured by the STEM detector, it is essential to use correct geometrical parameters of the used detector together with working distance of a sample in view of system pole piece - sample - detector. Those characteristics of our system are shown in the **Table 1**. The MC simulation was performed in Casino software [4] with appropriate settings (beam energy 30 keV, 100,000 electrons per point, total and partial cross section taken from Elsepa database, supporting thin carbon layer was taken into account). Captured electron microscopy images were calibrated to full and blanked electron beam by the equation (1)

$$I_{NORM} = \frac{I_{SIG} - I_{DARK}}{I_{FULL} - I_{DARK}} \tag{1}$$

where:

I_{SIG} - the pixel value

I_{DARK} - the mean value of dark image

I_{FULL} - the mean value of full electron beam on the detector.

Table 1 Radius and angle of individual detector segments of STEM III detector in working distance of 4 mm. The segments used in this study are highlighted by gray color.

Segment of the detector	Radius (mm)		Angle (mrad)	
BF	0.00	0.46	0.0	32.5
DF1	0.53	0.79	37.8	52.0
DF2	0.80	1.03	57.3	73.7
DF3	1.11	1.45	79.1	103.1
DF4	1.52	2.21	108.4	156.3
HAADF	2.28	10.00	161.5	650.3

Used calibration images are shown in **Figure 1.** Segments DF3 and HAADF are only visible in our SEM when the retractable STEM detector is manually shifted. When those images were taken, the detector was centred according to the electron beam. Notice the dark spot in the middle of the BF segment of the detector - this response inhomogeneity of our detector has clearly visible impact on the captured BF image. Slightly scattered electrons passing through the edge of spherical nanoparticle are landing on more sensitive part of the BF segment and creating the bright rim of the particle. It is recognisable in final determination of thickness, where influence of this inhomogeneity is visible in wrong estimation of thickness of supporting carbon layer (3 nm in thick).

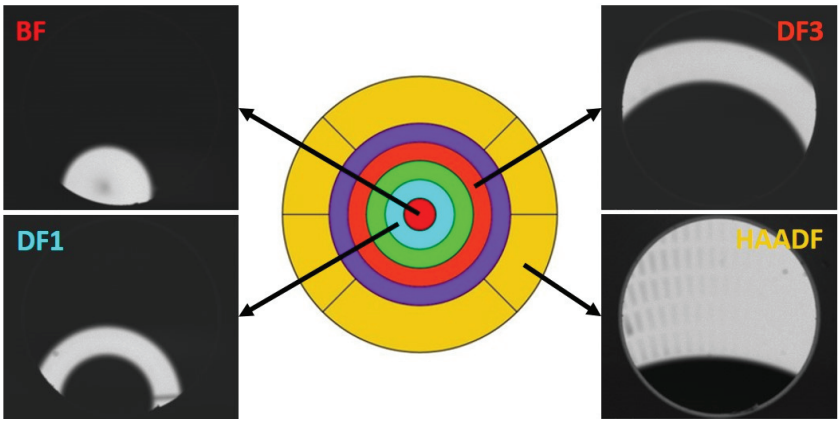

Figure 1 Calibration images of used detector segments with schematic drawing of their arrangement in used STEM detector

To compare individual detector segments and their accuracy we used - analogically to our previous work with a BSE signal [5] - a sample with a thickness known in each point - Latex nanosphere with a nominal diameter of 616 nm (S130-6, Agar Scientific, United Kingdom). We measured diameter of 583 nm in the case of the shown particle. Further processed images are shown in **Figure 2.** Even with the naked eye, there is low contrast change of DF3 image visible in direction from the edge of the particle to the centre. For MC simulation, the used density of latex was 1.055 kg/m^3. Results of MC simulation are shown in **Figure 3 right**.

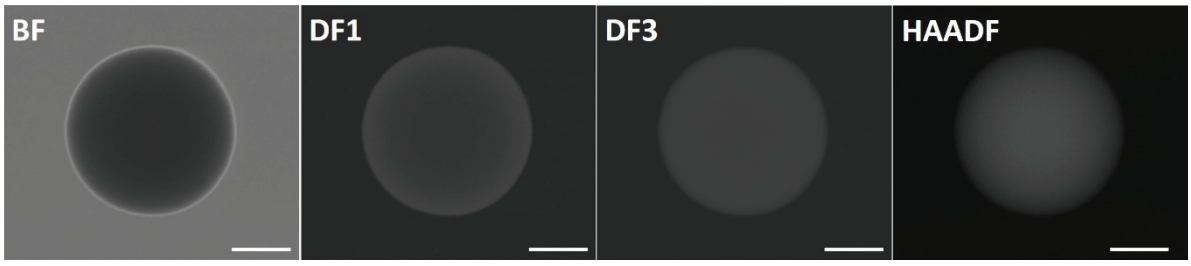

Figure 2 Latex nanosphere captured simultaneously in BF, DF1, DF3 and HAADF. Bar 200nm.

For noise limitation and easier comparison of the results, the center of nanoparticle is detected and then rotation averaging is performed (shown in **Figure 3**). In this step a distance of each individual pixel to center of the sphere is calculated. Then the pixels with distance in chosen range are averaged. The advantage of this method is the absence of an interpolation error. However, a small amount of pixels in the middle of the particle causes significantly higher noise in this area.

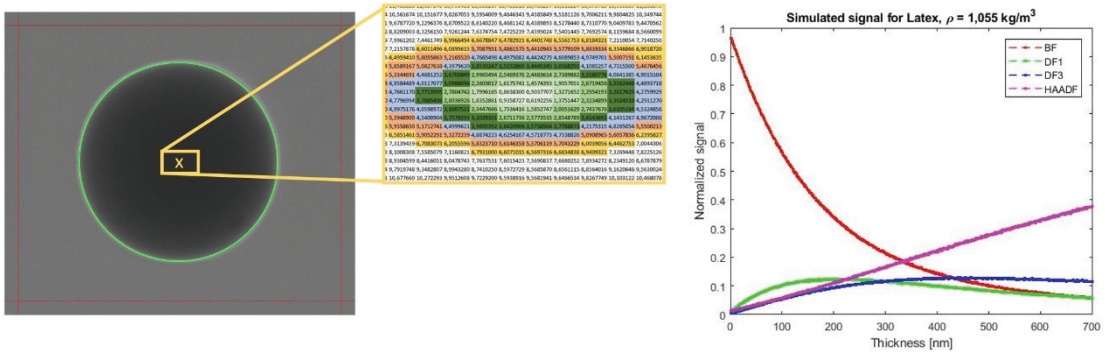

Figure 3 Left: Calculation of rotationally averaged signal. Detected sphere nanoparticle is green, control of integrity of the particle (red lines) and detected center (yellow cross). Right: Monte Carlo simulation for Latex.

3. RESULTS AND DISCUSSION

Our results show high differences of accuracy in estimated thickness by individual detector segments. Captured data are shown in **Figure 4 left** and final correspond thickness profiles are shown in **Figure 4 right**. The highest accuracy we found in the case of HAADF segment, where the estimated thickness corresponds with the theoretical one in the whole measured thickness range. The BF segment brings similar sensitivity as HAADF does, except for the center of the sphere and the edge area of the sphere including the thin support carbon layer. This inaccuracy is caused by reduction of sensitivity in the center of the BF segment of the used STEM detector (as mentioned above and shown in **Figure 1**).

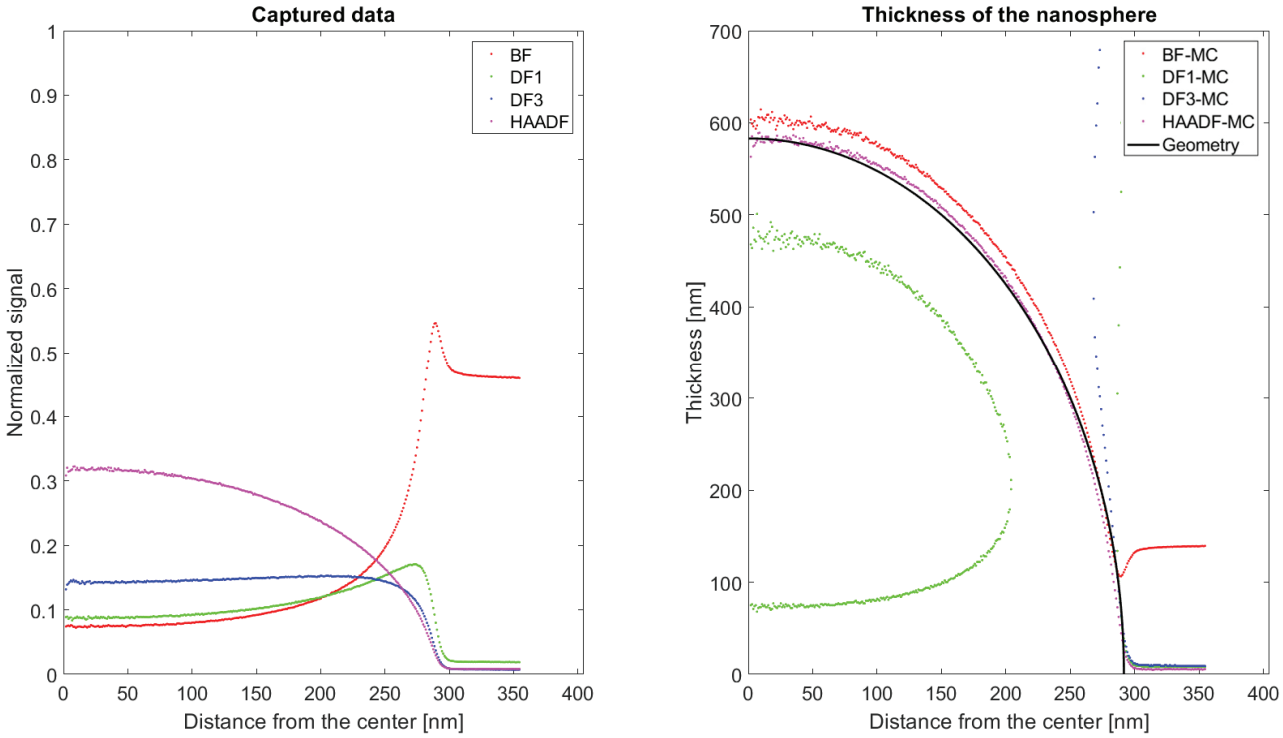

Figure 4 Captured data after rotationally averaging (left). Thickness of the sample estimated by quantitative STEM imaging in four different detector segments (right).

Results of DF1 and DF3 segment imaging have not been satisfying within our experiment so far. It is for further examinations, it is highly probable the calibration of both segments may be problematic. In the case of DF1 (green) there is dependency similar to what was expected clearly visible but moved about 100 nm lower in thickness. The straight line indicates the thickness over simulated range (values of the points where captured signal is higher than the highest simulated signal are removed). It brings the assumption, that the active size of those detector segments is higher than we used for MC simulation and our information about properties of used STEM detector is not accurate. The DF3 segment is not the appropriate choice for thickness estimation of our test sample, because of flat course at most of the captured data profile. Low change of signal due to varying thickness brings low sensitivity in thickness determination. In general, all segments DF1-DF4 have very fine angular range and thus their sensitivity for measured samples is very low.

4. CONCLUSION

We demonstrated the ability of quantitative low voltage STEM imaging to precise determination of local thickness in wide range from 0 to approximately 600 nm in the case of latex (polystyrene) sample. The proof was done at the sample of latex spherical nanoparticles that have relatively low density. Particles are

composed of light elements - carbon and hydrogen. In the case of samples containing higher atomic elements, the penetration depth of the electron beam is much lower. Unfortunately, not all used segments gave us the same results. We observed calibration errors which were devaluating measurements in DF1 and DF3 segments. Compared to that, BF and HAADF segments bring accurate results which correspond with the reality of the sample. Nowadays other experiments are underway with the aim to observe origin of partly inaccurately calibration. We recommend to process similar comparative experiment after any change in imaging process of a microscope.

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