Automated quantification of 2D and 3D STEM spectrum image data including thousands of nanoparticles

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The structural morphology and elemental distribution of nanoparticles are essential to their performance, in applications such as catalysis and optical labeling[1 - 3]. Particularly, multi-element nanocrystals provide the opportunity to engineer performance through combining properties of two or more elements. Scanning transmission electron microscopy (STEM) and X-ray energy dispersive spectroscopy (EDS), combined with recent advances in electron tomography, enable both two dimensional (2D) and three dimensional (3D) characterisation of the nanoparticles' structure and composition[1,4 - 6]. However, populations of nanoparticles very often possess large deviations in size and composition. The majority of specimens investigated by STEM either lack truly statistical sampling, or require time-consuming manual analysis. When moving from 2D to 3D datasets, acquisition times and data size increase dramatically, up to 140× (±70° tilt range and 1° increment), as does the time required to analyse data.

The time constraints and under-sampling for analysing both 2D and 3D nanoparticle datasets are overcome by our high-throughput processing methodology. The approach consists of automated image processing, particle segmentation and machine learning based analysis. We apply the approach to two cases that involve thousands of nanoparticles characterised by STEM high-angle annular dark field images and EDS; 2D images of bi-elemental hollow nanoshell catalysts and 3D data of multi-elemental nanorods. Due to an order of magnitude increase in specimen sampling, we were able to investigate the heterogeneous nature of both specimen populations. On the 2D data, surface roughness and size of each nanoparticle is analysed with 0.5 nm precision[7]. The 3D data reveals preferred doping positions for multiple elements on each nanoparticle. The analysis of heterogeneity, correlated with synthesis conditions, provides feedback to chemists for better control of nanoparticle synthesis. The shared features such as chemical segregation sites improve the understanding of nanoparticle performance mechanisms. We also anticipate the automated methods developed in this study can be extended to any nanoparticle population, regardless of its application.

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