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Automated Image Processing Scheme to Measure Atomic-Scale Structural Fluctuations

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The interaction of gases with a solid catalyst nanoparticle during catalysis is a non-equilibrium process that requires high spatial and temporal resolution measurements to elucidate underlying mechanisms. State-of-the-art environmental transmission electron microscopy (ETEM) enables *in situ* measurements of the dynamic changes occurring under reaction conditions [1,2]. These changes usually take place rapidly at the nanometer scale. Recently, direct electron detection cameras, have enabled us to record atomic-resolution images with ns time resolution, but generate videos with large amount of data (≈ GB s^{-1}). It is laborious to manually analyze such large-size videos, frame by frame, to extract the events of interest at the required time resolution. Automated analysis would be preferable, but is complicated by (a) noise in individual frames due to rapid readout times and (b) sample drift that occurs in a single video recording period. In order to overcome these issues, we have developed an automated image processing scheme (AIPS), to obtain structural information from the images extracted from videos. AIPS uses a combination of algorithms publically available and developed at NIST that perform noise reduction, drift correction, template matching, atom-position location, and triangulation to accurately determine the positions of atomic columns. We tested our method by quantitatively relating the crystal structure fluctuations in a catalyst nanoparticle to the growth of single-walled carbon nanotube (SWCNT) as a function of time.

We have employed an environmental transmission electron microscope (ETEM), operated at 300 kV with an aberration corrector, to record real-time atomic-resolution videos (at a frame rate of 6 s^{-1}) of SWCNT growth from a Co-Mo/MgO system in C_2H_2 gaseous environment at 650 °C [3]. Two distinct regions, R1 and R2, within the catalyst particle can be visually identified (Fig. 1a). Fast Fourier transform (FFT) analysis of these regions revealed them to be Co and Co_2C, respectively (Fig. 1b and 1c). However, the total area of the two regions fluctuated with time and FFT analysis was not always possible. AIPS enables a real-space analysis that can be used to determine the area occupied by different structures at any given time (Fig. 2a). For each atom-column position, the distances to the neighboring atom columns were averaged, giving a local spacing value which was used to determine whether a local region was Co or Co_2C (Fig. 2b). A simulated TEM image (Fig. 2c), using a structure model based on AIPS analysis, was used to assign colors to different structures for visualization purposes (Fig. 2d). The structural gradient within the particle along the red arrow marked in Fig. 3a confirmed that the region enclosed within the growing SWCNT was Co rich while the outer few layers were mostly Co_2C (Fig.3b). The histogram of measured spacings (Fig. 3c) has a sharp peak centered at ≈ 0.22 nm and another broad peak centered around ≈ 0.25 nm. The Gaussian fit of the first and second peaks were found to have a standard deviation of 0.007 nm (7 pm) and 0. 015 nm (15 pm), respectively and an uncertainty of 0.01 nm (10 pm). While the first peak can be unambiguously assigned to Co, the second peak may be due to co-existing Co_3C (0.25 nm) and Co_2C (0.243 nm). Additionally, the fluctuation of C content in the nanoparticle was compared with the SWCNT growth length. We found that when the Co fraction increases – carbon output is greater than carbon intake – the tube growth increases. Similarly, when the Co fraction decreases – carbon intake is more than carbon output − the tube growth rate decreases. We show AIPS not only reduces the analysis time but also enables us to

make quantitative measurements of reaction processes. The presentation will also include the application of this methodology to other reactions such as the reduction rate of α-Fe_2O_3 nano-blades.

References:

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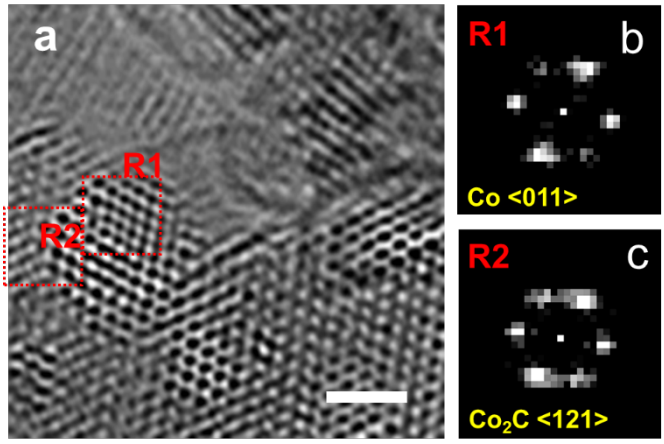

Figure 1. (a) A high resolution image of SWCNT growth from a catalyst particle that contains two different structures (R1 and R2) identified as Co and Co_2C, respectively using (b, c) FFT analysis.

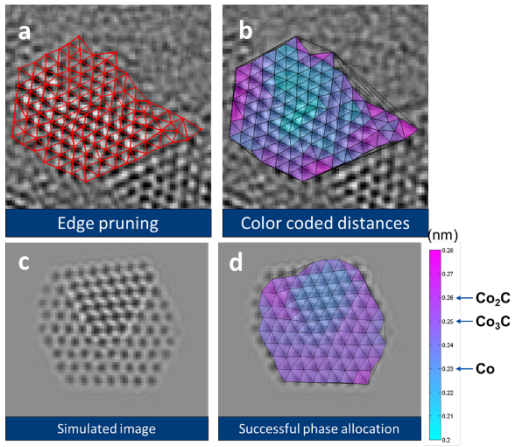

Figure 2. (a) Atomic positions and distances overlaid in red, (b) distribution of different phases within the particle, (c) simulated HREM image using Co and Co_2C structures, coded as blue and magenta respectively in (d)

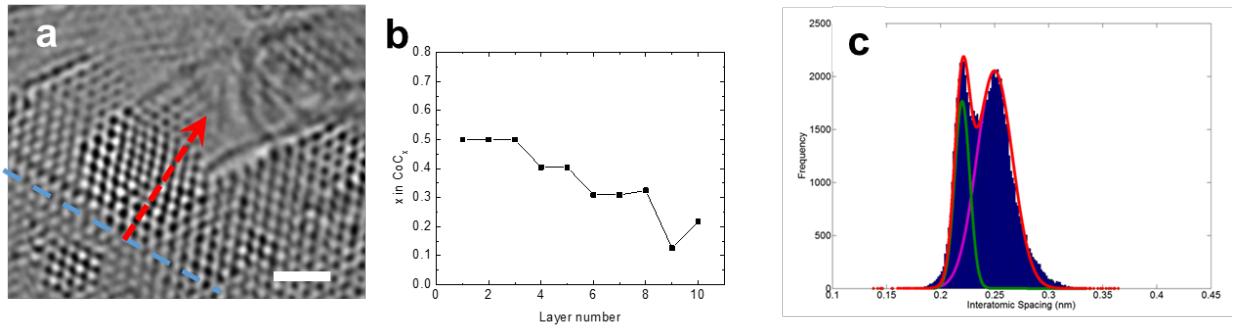

Figure 3. (a) Direction of the structural change is marked by the red arrow, (b) measured carbon content in individual layers decreases in the direction of the arrow, showing that a Co-rich region is enclosed in the SWCNT, (c) a histogram of local structural parameters for all triangles (Fig. 2a) in all frames of the video showing a bimodal distribution of interatomic spacings, representing Co and Co_2C structures, respectively.

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