Towards Quantitative Aberration-Corrected Monochromated Environmental High-Resolution Transmission Electron Microscopy of Dynamic Processes

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As a result of recent instrumental advances, it is now possible to combine aberration correctors and monochromators with the ability to examine specimens in the transmission electron microscope in the presence of external stimuli, such as exposure to reactive gases, electrical currents and illumination with light. This new parameter space provides a wealth of interesting opportunities, but also an overwhelming temptation for users of instruments such as aberration-corrected "environmental" transmission electron microscopes (ETEMs) [1, 2] to proceed rapidly with atomic resolution studies of chemical reactions and growth processes. However, considerable care is required, both because a quantitative description of the effect on images, diffraction patterns and electron energy-loss spectra of electron scattering from gas atoms in the highest-pressure part of the electron column is not yet available, and because of significant differences between dynamic experiments carried out in an electron microscope from similar *ex situ* studies. In particular, the effect of the incident electron beam on damage processes, on ionization of the gas and the specimen and on chemical reaction rates needs to be considered, even at room temperature.

We have recently installed an FEI Titan 80-300 ETEM equipped with an objective lens aberration corrector, a monochromator and a differential pumping system (Fig. 1). Seven gases (H_2, He, CH_4, N_2, CO, O_2 and Ar) are connected permanently and can be introduced at pressures of up to ~1500 Pa. Figure 2 shows a high-resolution image of Au particles on a BN support recorded in the presence of 320 Pa of O_2, while Fig. 3 shows images taken from an *in situ* study of the sintering of Au particles on a BN support in the presence of 130 Pa of H_2 at 400°C. Two sintering mechanisms have been proposed, based on i) migration of particles and coalescence when they are in close proximity and ii) mass transport from smaller to larger particles by the diffusion of atoms or molecular species (i.e., Ostwald ripening). Inspection of Fig. 3 shows that particles 1 and 2 coalesce from a) to b). In b) and c), the same phenomenon is observed for particle 3. From c) to f), particle 4 becomes smaller, suggesting that Ostwald ripening is taking place. These images highlight the fact that both mechanisms can take place under the same conditions. The combination of *in situ* capabilities and a monochromated electron source allows for the acquisition of electron energy-loss spectroscopy (EELS) data with improved energy resolution during chemcial reactions (Fig. 4). It is also important to note that the intensities of recorded images decrease with increasing gas pressure (Fig. 5) and that scattering of primary electrons by gases results in a loss of image resolution, the more so for heavier gases. These effects must be considered when analyzing images [3, 4].

References

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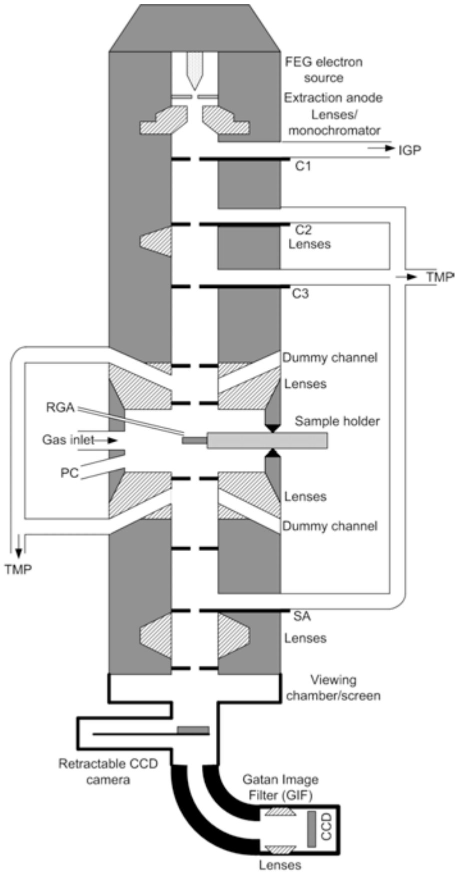

FIG. 1. Schematic diagram of a differentially pumped TEM column. FEG: field emission gun; IGP: ion getter pump; TMP: turbo molecular pump; RGA: residual gas analyzer; PC: plasma cleaner; C1: first condenser aperture; SA: selected area aperture.

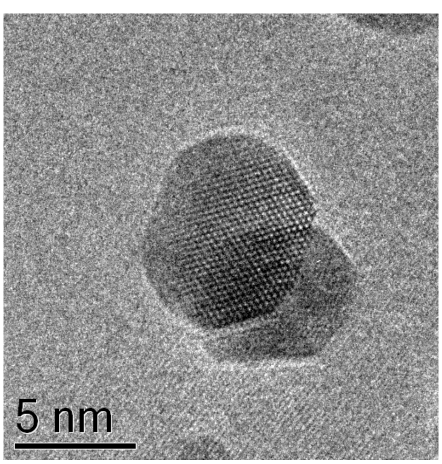

FIG. 2. High-resolution TEM image of Au particles on a BN support imaged in 320 Pa of O_2 at room temperature.

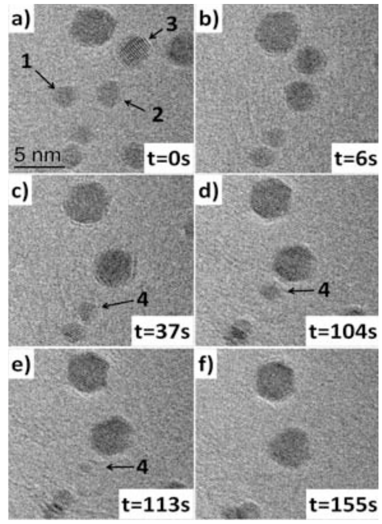

FIG. 3. Au particles on a BN support imaged in 130 Pa of H_2 at 410°C at the indicated times at 300 kV with C_S set to below 5 μm. The numbers refer to particles that undergo different sintering mechanims.

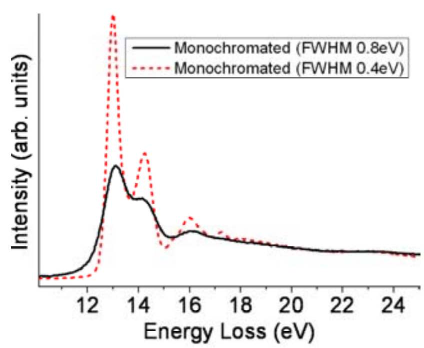

FIG. 4. Non-monochromated and monochromated EELS spectra acquired in an ETEM from N_2 gas.

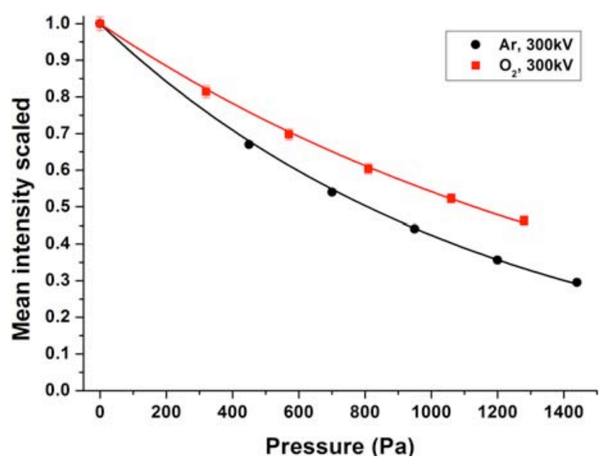

FIG. 5. Normalized image intensity in vacuum (with no specimen present) measured as a function of gas pressure for Ar and O_2 at 300 kV.