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Quantification of carbon contamination under electron beam irradiation in a scanning transmission electron microscope and its suppression by plasma cleaning

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Abstract. We have measured the build-up of carbon surface contamination as a function of time and irradiated area size for various specimens in a JEOL 2010F (scanning) transmission electron microscope, employing both t/λ mapping with our Gatan imaging filter and recording changes in annular dark-field image intensity. It is shown that the total number of carbon atoms deposited per time for a given beam intensity is roughly constant at room temperature for as-received specimens while it is significantly lower for plasma cleaned specimens. This explains why contamination is generally only an issue at the highest magnifications where the contamination regions become smaller and the carbon layers correspondingly thicker. A Fischione plasma cleaner was then used to remove these carbon layers, and the rate of carbon removal has been determined for contamination spots produced in stationary spot mode as well as for extended regions scanned for a minute so that optimal cleaning times can be chosen.

1. Introduction

The quantitative analysis of scanning transmission electron microscopy data, images as well as spectra, is often hampered by surface contamination during electron irradiation. The gradual build-up of carbon layers on the specimen will reduce the image contrast and increase the signal from C K-edge energy losses and corresponding X-rays, thereby causing artefacts in spectroscopy.

Contamination is caused by the cracking of hydrocarbons present both in the vacuum chamber and on the specimen and their subsequent surface diffusion to the site being irradiated [1,2]. Although our JEOL2010F field-emission gun (scanning) transmission electron microscope (FEG-(S)TEM) is equipped with a completely dry vacuum pumping system that involves only scroll, turbo molecular drag and ion getter pumps but no rotary or oil diffusion pumps, carbon contamination is still an issue when imaging or scanning small regions at high magnifications. For this study, the microscope has been tested just before a bake-out of its column was due to improve vacuum levels. We have calculated quantitatively the build-up of carbon surface contamination as a function of time and irradiated area size for different specimens and holders. A Fischione Model 1020 high-frequency argon/oxygen plasma cleaner was then used to remove these carbon layers, and the rate of carbon removal has been determined so that optimal cleaning times can be determined.

2. Experimental

Samples of C, InAs and InGaAs were investigated in the JEOL 2010F FEG-(S)TEM in scan or spot mode at nominal magnifications between 60kX and 4000kX (4MX), using an electron beam ~0.5nm in diameter and with ~0.1nA current (nominal spot size M, 100-130μA emission, 197kV). The

electron beam was either held stationary (spot mode) or raster scanned over square regions (scan mode). In order to facilitate quantitative evaluation, usually several frames of 1min exposures were taken at successively smaller magnifications so that the regions scanned at e.g. 4MX times were then imaged at 1MX times, 250kX and 60kX, revealing a superposition of the corresponding carbon contamination layers, almost square in size if drift was small, in the final image.

In high-angle annular dark field (ADF) imaging, the signal detected depends on the product of the square of the average atomic number of the atoms ('Z-contrast') and the number of atoms (i.e. the thickness) [3], and carbon contamination deposited on any of the free surfaces of a sample will show up as an increase in intensity and a reduction of image contrast from the underlying original specimen material. Figure 1 depicts such a typical ADF image taken at 120kX of an area scanned before at higher magnifications, where the contamination windows produced by the previous scans are clearly visible. From such an image we can measure the sizes of the contaminated areas, as in Figures 1 and 2. Figure 3 shows that at liquid nitrogen temperature carbon is etched away instead of being deposited, for the same sample as before. The inner collection angle for ADF imaging in all cases was ~40mrad.

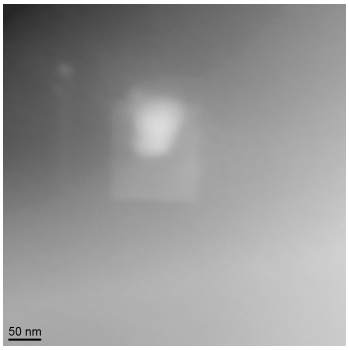

Figure 1. ADF-STEM at 120kX magnification of contamination windows on InAs produced during prior scanning for 1min each at magnifications of 2MX and 500kX. Room temperature

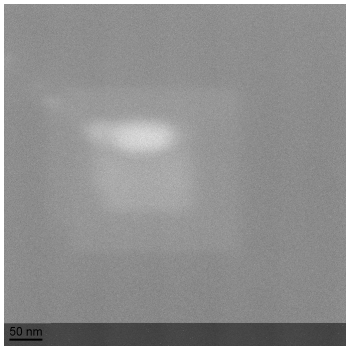

Figure 2. ADF-STEM at 120kX magnification of contamination windows on C test specimen produced during prior scanning for 1min each at magnifications of 1MX, 500kX and 250kX. Room temperature

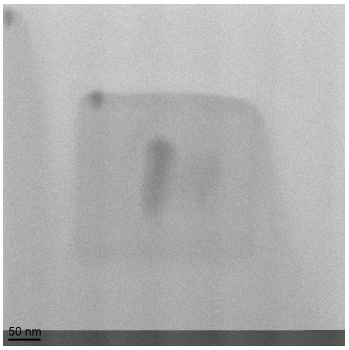

Figure 3. ADF STEM at 120kX magnification of contamination windows on C test specimen produced during prior scanning for 1min each at magnifications of 1MX, 500kX and 250kX. T= –173 ºC (100K)

3. Results and Discusssion

Figure 4 depicts t/λ maps from an area of InAs with carbon contamination areas and contamination spots after several periods of plasma cleaning at standard conditions (25% O_2, 75% Ar mixture, 13.56MHz, no shield). The gradual disappearance of the carbon contamination can be clearly monitored. λ denotes the inelastic mean free path of the material, which is a measure of the degree of inelastic scattering. As scattering probabilities add up, for several materials on top of each other the mean free paths are additive, i.e. for a stack of i different materials $t/\lambda=\Sigma\, t_i/\lambda_i$ [4]. Absolute thicknesses of carbon overlayers on InAs can thus be determined if the inelastic mean free paths of both materials are known ($\lambda_C\approx$130nm and $\lambda_{InAs}\approx$84±13nm for U=197kV, β=40mrad [4]) and if the InAs thickness is measured. The latter can be determined indirectly by ADF imaging of the clean InAs region at the beginning of the experiment, assuming the intensity scales linearly with the product of thickness, t, and mean atomic number, Z, to the power of some exponent ε: $I_{ADF}-I_0=ctZ^{\varepsilon}$, where I_0 is the dark current signal offset (background in vacuum without specimen or beam off) and c=const. The proportionality constant can be determined for given ε. For an inner collection angle β=40mrad we used $\varepsilon\approx$1.6, based on earlier experiments studying ADF image intensities quantitatively as a function of collection angle [5].

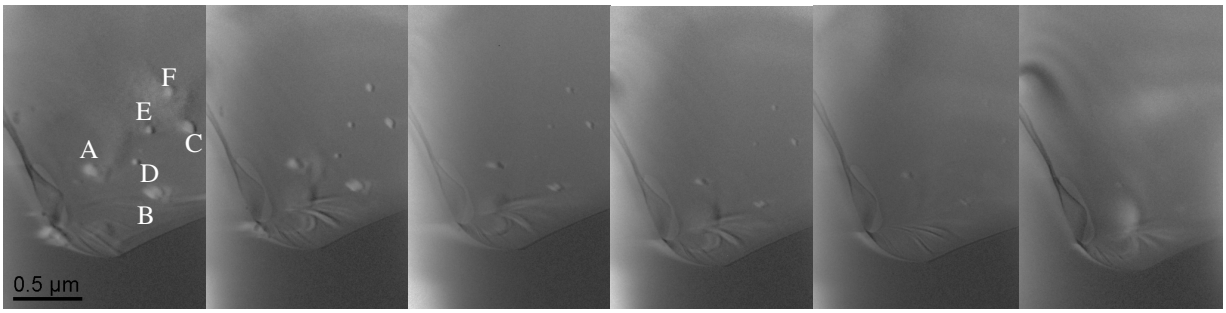

Figure 4. Relative thickness maps obtained of the same InAs area after successive plasma cleaning. The leftmost image shows the condition at the start (t=0), the series from second left to right shows maps after 1, 2, 4, 6 and 8 minutes of cleaning. The labels indicate the positions of the contamination windows produced in scan mode (A-C) or pillars (D-F) in spot mode, respectively.

The effectiveness of the plasma cleaner is demonstrated in Figure 5, which plots the carbon thickness, d, thus determined on top of the InAs samples as a function of time, t. The relationship is clearly exponential and can be fitted by functions of the form $d=d_0 \exp(-\lambda t)$, with time constants of λ^{-1}~2min for flat contamination windows produced during scanning and ~4min for contamination pillars produced in stationary spot mode. We explain this by the difference in aspect ratios: contamination spots have the form of pillars and thus offer a smaller surface for the plasma to etch away the carbon, compared to flat carbon films that have a larger free surface. Our findings are in agreement with previous studies on the same plasma cleaner that indicated 2min [6] to 5min [7] would suffice to remove surface C contamination almost completely.

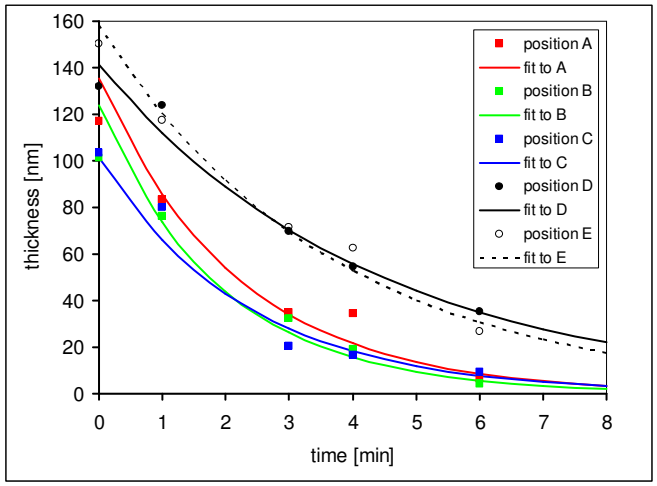

Figure 5. Plot of thickness of remaining carbon deposit as a function of plasma cleaning time. Square symbols (A, B, C) are for flat contamination windows produced in scan mode, round symbols (D, E) represent contamination pillars obtained in spot mode.

position	t_0 [nm]	λ [1/min]	R
carbon films (scan mode)			
A	135.3	0.4596	–0.9665
B	123.8	0.5178	–0.9843
C	101.2	0.4290	–0.9796
average	120.1	0.47±0.05	–0.9768
carbon pillars (spot mode)			
D	141.4	0.2323	–0.9938
E	157.9	0.2741	–0.9863
average	149.7	0.25±0.03	–0.9901

Table 1. Statistical evaluation of contamination data from Figure 5. t_0= initial thickness ; λ=inverse time constant for exponential fit; R=regression coefficient for exponential fit

The volume of carbon contamination thus determined from the product of lateral extension along x and y directions and thickness can be converted into atomic numbers, given a molar volume of 5.29cm^3. The number of carbon atoms deposited while keeping the beam scanning for 1min at a given magnification is plotted in Figure 6 and indicates, for uncleaned InAs or InGaAs at room temperature, values around 1-3×10^8 C atoms for both specimens. For a beam current of 0.1nA this implies that ~200 electrons are needed for depositing a single carbon atom. This observation may be explained by

the total charge of the electron beam being the relevant parameter for cracking of hydrocarbon bonds. Thus, scanning n-times smaller areas at n-times higher magnifications will lead to n^2 times thicker carbon layers, which explains the importance of the effect only at highest magnifications. Plasma cleaning the sample for 8min reduces contamination by a factor of ~20, in agreement with [7], i.e. ~4000 electrons would deposit a single carbon atom. An amorphous C film cooled by liquid nitrogen to ~100K, however, is gradually etched away, as reported in [8], and it is interesting to note the etching rate decreases with magnification, as shown in the right part of Figure 6. This confirms the rôle of the free surfaces from where carbon atoms are lost by knock-on processes.

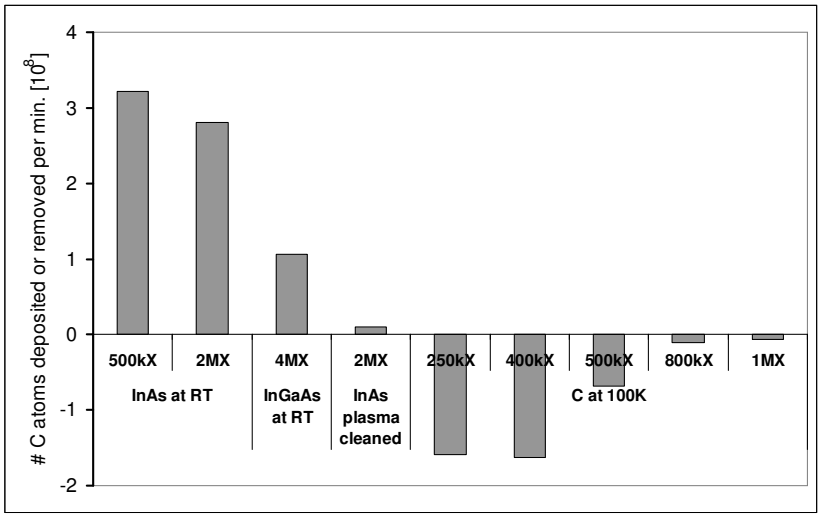

Figure 6. Plot of total number of carbon atoms deposited (positive sign) or etched away (negative sign) by electron irradiation in our JEOL 2010F during 1min scan mode with medium sport size at various conditions (RT=room temperature). Note the only small difference between different specimens (InAs vs. InGaAs), but the large influence of magnification, prior plasma cleaning (for the InAs sample) and specimen temperature.

4. Summary

It has been shown that scanning a small electron beam in FEG-STEM experiments leads to surface carbon contamination, the rate (volume of carbon per electron dose) of which is almost constant. In our JEOL2010F microscope, just before bake-out, ~200 electrons at 197kV yield one C atom of contamination for a normal argon ion milled sample, while plasma cleaning the sample for 8min reduces contamination by a factor ~20. For a given specimen cleanliness, a constant contamination rate explains why carbon contamination is only an issue at high magnifications, as the area scanned is then very small and the contamination film correspondingly thicker. Plasma cleaning reduces the amount of carbon contamination on the surface exponentially as a function of plasma cleaning time, with a time constant for the decay of the remaining carbon thickness of 2-4 minutes.

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