

Electron-beam-induced surface diffusion of contaminants and growth of carbon contamination

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Background incl. aims

Carbonaceous contamination deposited on samples illuminated by a beam degrade the quality of images and aggravate structural and compositional analysis of the material. Contamination results from irradiation-induced polymerization of contaminants, which can be hydrocarbon molecules already present on the sample surface or molecules adsorbed from the residual gas of the instrument chamber [1]. Several authors concluded that the contribution of surface contaminants is dominant [2,3], but already earlier models consider an additional source from the residual gas in the instrument [4].

The goal of this work is to describe the flow of contaminants and their polymerization in the irradiated area with a number of parameters reduced to a minimum necessary. To analyze possible sources of contaminants, both, a surface diffusion process and the supply of contaminants from the residual gas are considered. It is hypothesized that the diffusion process is driven by the gradient of the surface density of contaminants, generated by the impact of the electron beam probe. The contribution of the residual gas atmosphere in the instrument is described by the tendency to re-establish an equilibrium surface density of contaminants.

A time-dependent reaction-diffusion model is elaborated to describe contamination growth on surfaces illuminated by an electron beam. The unknown parameters of the model are determined by comparing the theoretical predictions with experimental results. The experiments are designed such that the influence of each parameter can be unequivocally separated. Successive contamination measurements, performed at distinct time intervals allow to follow the dynamic of the process. Examination of the individual contribution of contaminants from the sample surface and from the residual gas of the instrument is facilitated by experiments with appropriate variations of these parameters.

Methods

Circular contamination patterns were grown in a FEI DualBeam Strata 400S on commercial thin (~10 nm) amorphous carbon films by illuminating the sample homogeneously with a defocused electron beam. The radius of the irradiation disk was chosen to be sufficiently large (~ 700 nm) to separate the influence of the distinct parameters on the contamination growth. Thus, the inflow of the surface contaminants by diffusion was noticed at the margin of the disk, while the residual gas contributed on the entire sample surface. The dynamical evolution of the process was followed by successive contamination measurements performed up to 20 minutes at time intervals of 5 minutes.

The local height and shape of the contamination rings were quantified by taking high-angle-annular-dark-filed (HAADF) images in scanning-transmission-electron-microscopy (STEM) mode between the irradiation cycles, and comparing their intensities with corresponding Monte-Carlo simulations. Both, irradiating and imaging, were conducted at electron beam energy of 20 keV and with a current of 120 pA.

To elucidate the supply of contaminants from the residual gas, the chamber pressure of the microscope was varied by different pumping periods and the usage of a cooling vessel trap.

The initial density of the surface contaminants was altered by subjecting the sample to variable duration of in-chamber plasma cleaning.

Consideration of the main parameters of the process lead to a reaction-surface-diffusion model with an additional source term. It was supposed that the relative density of contaminants $n = N/N_0$, normalized to an initial density $N_0 = N(t = 0)$, changed locally due to polymerization to amorphous carbon contamination by the illuminating beam. The occurring gradient of n generated a surface diffusion process of contaminants, characterized by the diffusion coefficient D . The reaction of the irradiating electrons with the contaminants was described by the reaction-frequency σ . An adsorption frequency η of molecules from the residual gas considered for a source term with the tendency to re-establish the initial density of the surface contaminants. With these terms, the reaction-diffusion equation was elaborated as follows:

$$\partial n / \partial t = D \cdot \Delta n - \sigma \cdot n + \eta \cdot (1 - n),$$

with Δ representing the Laplace operator in cylindrical coordinates for radial symmetry. Comparing the predictions of this model with the time evolution of the experimentally grown contamination, the parameters of the process could be determined. All results were considered with respect to reference measurements, performed prior to the experiments with parameter variation.

Results

A surface-plot in Fig.1 a) shows the HAADF-STEM image intensities of contamination grown during 20 minutes of local irradiation with a defocused electron-beam of 20 keV. The modeled thickness profiles (continuous lines) in comparison with the measured radial profiles (dashed lines) of the contamination ring are depicted for distinct time intervals in Fig.1 b). Comparing the predictions of the reaction-diffusion model with experimentally grown contaminations, the diffusion of the surface contaminants was described by $D \sim 10^3 \text{ nm}^2/\text{s}$ and the polymerization of the contaminants by a reaction frequency $\sigma \sim 1 \text{ Hz}$. Small variations of D and σ for distinct measurements could be explained by local differences of the contaminants type of the corresponding experiments.

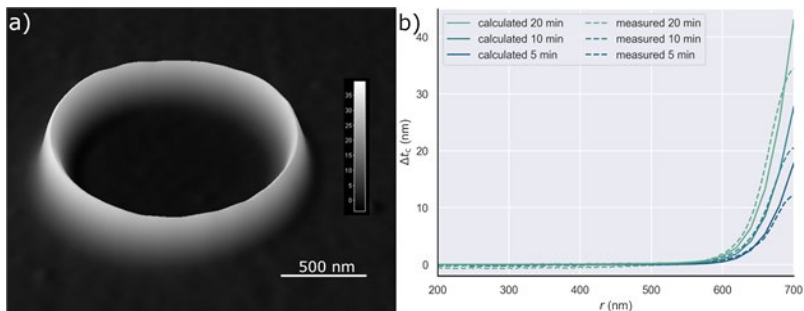
The contribution of contaminants from the sample surface and from the residual gas were separately determined by varying accordingly the experimental conditions by plasma cleaning, respective pumping and cooling of the instrument chamber. Already short plasma cleaning durations significantly reduced contamination growth and the exponential decay in time of surface contaminant density N_0 showed an effective removal of surface contaminants.

With higher chamber pressures, the residual gas supply of contaminants increased, as indicated by the raise of the adsorption frequency of contaminants. However, its value $\eta \sim 0.002 \text{ Hz}$ remained very low in comparison with the other parameters and contributed only with a small amount to the total contamination growth. Its direct influence in the illuminated area was relatively reduced, but became noticeable for contaminant adsorption outside the illuminated area and their subsequent diffusion into it.

Conclusion

In conclusion, it can be asserted that contaminants stem from the sample surface and diffuse into the illuminated area. This underlines the effectiveness of in situ plasma cleaning. The residual gas contributes less to direct contamination growth but should be considered for experiments of longer duration.

Graphic:



Keywords:

contamination, surface diffusion, electron microscopy

Reference:

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