Micron 196–197 (2025) 103857

Contents lists available at ScienceDirect

Micron

journal homepage: www.elsevier.com/locate/micron

Electron irradiation-based cleaning of the scanning electron microscope and its samples

András E. Vladár

National Institute of Standards and Technology, Gaithersburg, MD 20899, USA

ARTICLE INFO

Keywords:
Electron irradiation cleaning
SEM
Sample
Contamination

ABSTRACT

The deposition of carbonaceous material under electron beam irradiation is an old and persistent problem in scanning electron microscopy. It impedes high-resolution imaging and measurements, especially at the nanometer scale. The emergence of contamination is a complex process of adsorption, dissociation, and desorption of carbonaceous molecules. Depending on the kind and amount of precursor molecules, vacuum, sample material, temperature, and the intensity and energy of the irradiating electrons, deposition can overwhelm removal or the other way. Fortunately, with the introduction and commercial availability of low-energy, plasma-based, and other cleaning devices, contamination can be reduced to non-detectable levels. Plasma devices, working in low vacuum, which is necessary to start and sustain plasma generation, can effectively remove contamination precursor hydrocarbon molecules to the extent that a clean sample can be imaged and continuously measured for hours without deposition of any perceptible amount of carbonaceous contamination. Here, we report on the results of a new and effective cleaning device that has recently become available, which is different from plasma cleaning devices. It is based on low-energy electron irradiation and works in high and ultra-high vacuum (UHV).

1. Introduction

Focused-electron-beam-induced deposition (FEBID) on one side is a valuable technique for nanometer-scale fabrication of two- and three-dimensional structures of various materials with potential applications in integrated optical devices, photolithography mask fabrication, and repair (Perentes et al., 2004; Lianga et al., 2005; Fowlkes et al., 2018). FEBID, on the other hand, by deposition of carbonaceous contamination, can be a scourge in electron microscopy, with its effects ranging from nuisance in measurements to devastation of the sample.

Traditionally, a clean sample surface was required only for actual surface quantitative measurement techniques like Auger electron spectroscopy, electron spectroscopy for chemical analysis, and secondary ion mass spectrometry, which work in UHV to achieve and maintain clean sample surfaces. Clean sample surfaces are also essential in ultra-high-resolution microscopy, low-landing energy microscopy, and compositional analysis, especially when long dwell times or high beam intensities are required, e.g., as mapping mode operation in electron energy loss spectroscopy, x-ray analysis and spectroscopy, electron backscatter diffraction and or when measurements of low concentration constituents are performed.

In the past, with high-vacuum (HV) systems, achieving and maintaining such high sample cleanliness has been impossible that it would not interfere at all with imaging and measurements, even in so-called clean, oil-free vacuum systems. Introducing low-energy plasma-based SEM and sample cleaning solutions has significantly improved this situation (Vladár et al., 2001). Today, it is possible to realize the state of ultra-high cleanliness (UHC). In the UHC state of a high-vacuum system, hours-long, continuous imaging or measurement of samples can be carried out without the deposition of any noticeable carbonaceous contamination (Toth et al., 2009). Maintaining the UHC state in HV systems requires proper management of both the instrument's vacuum and the sample surface cleanliness before and during the imaging or measurement processes.

The deposited contamination depends on the dose ($D=I_p*t$, i.e., the current of the primary electron beam multiplied by the irradiation time) and the abundance of precursor molecules at the surface. It also depends on the energy and number of low-energy electrons, mainly the so-called

Contribution of the National Institute of Standards and Technology; not subject to copyright in the United States. Specific commercial equipment, instruments, or materials are identified in this report to specify the experimental procedure adequately. Such identification is not intended to imply recommendation or endorsement by the National Institute of Standards and Technology or the United States Government Publishing Office, nor is it intended to indicate that the materials or equipment identified are necessarily the best available for the purpose.

E-mail address: andras@nist.gov.

https://doi.org/10.1016/j.micron.2025.103857
Received 11 March 2025; Received in revised form 2 May 2025; Accepted 19 May 2025
Available online 20 May 2025
0968-4328/Published by Elsevier Ltd.

secondary electrons (SEs) that, by definition, have energies up to 50 eV. At room temperature, carbon-containing molecules' chemical bond dissociation energies fall from 4 eV to 5 eV (Blanksby and Ellison, 2003; Cao et al., 2017). These low-energy electrons crack the chemical bonds of precursor molecules that, in a high vacuum, can easily move on surfaces and turn them into molecules that stick to the irradiated surface. The higher energy electrons make it possible to remove and clean the SEM sample chamber and sample surfaces, as well as already deposited carbonaceous molecules. Type 1 SE or SE_1 is an electron directly created at the primary electron's impact point; these electrons give the highest spatial resolution in SEMs. SE_2 is a detected electron generated by a primary electron that is inelastically scattered in the sample within the escape depth of an SE. These electrons emerge from a much larger spot than type 1 SEs. High landing energies primary electrons generate many secondary electrons, but these are mostly deep in the bulk of the sample, where they cannot escape. At low landing energies, more type 2 SEs can escape, worsening the contamination deposition and removal balance.

Even a monoatomic layer of carbon on the surface of a bulk material significantly affects the SE yield and the energy and width of the SE peak intensity. According to (Cao et al., 2017), on a clean Cu sample, the highest SE yield is at about 300 eV primary beam energy, just above 2, but a single graphene layer lowers it to about 1.5. The energy distribution of SEs has a peak intensity of 1.5 eV and a full width at half the maximum value of 4.7 eV on clean Cu, but with a single graphene layer on top, these change to 4 eV and 10.9 eV, respectively.

Depending on the cleanliness of the SEM and its sample, four contamination results can appear, see Fig. 1.

The left image shows the case of a contaminated SEM vacuum chamber, while the right one was taken after cleaning the SEM vacuum chamber. The dose was varied by changing the irradiated area, i.e., the magnification. These images were taken on a clean scanning electron microscope contamination assessment reference sample (SEMCARS) (The Scanning Electron Microscope Contamination Assessment Reference Sample (SEMCARS) (SEMCARS) (SEMCARS)) contamination testing artifact with a landing energy of 1 keV and beam current of 43 pA at a horizontal field width (HFW) of 2.54 µm. The image file contains many parameters, including the net frame time of about 28 sec and pixel dwell time of 30 µs. The primary electron dose for the irradiated 2.54 µm wide image frame was about 1.46 nC. This means that about 9.1 billion primary and a similar number of secondary and backscattered electrons were at work across the image's 1131,520 pixels. For 1 pixel, on average, the dose was about 1.3 fC by about 8052 primary electrons, and for one nm^2 area, the dose was 0.33 fC and by about 2045 primary electrons. The dose and number of electron values are approximate and belong to a single image frame with the above parameters.

This SEM does not use an over-scan method to make the irradiated area somewhat larger than the recorded image's area to minimize distortions and contamination artifacts. The picture on the left was acquired on a clean sample with the SEM when its cleanliness allowed one image with the above parameters to be taken without signs of contamination becoming visible. Still, imaging at higher doses did develop various amounts of it.

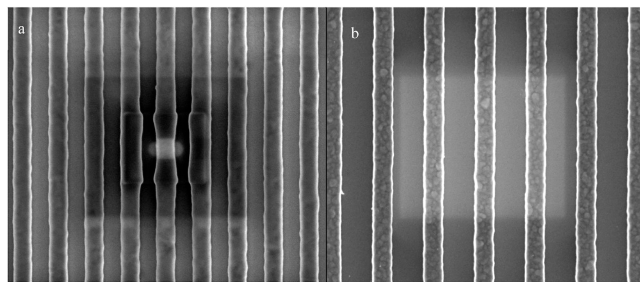

Fig. 1. Deposition of contamination depends on the dose and landing energy of irradiating electrons and the availability of precursor molecules. Image *a* was taken on a clean sample with a contaminated SEM, and *b* is after the SEM was cleaned using the NIST SEMCARS sample, a Si chip with amorphous Si patterns. Clean SEM and a clean sample make surface details more visible. 1 keV landing energy SE images acquired by the in-lens detector at HFW of 2.54 µm.

The SE image on the left was first irradiated at an HFW of 0.635 µm for 10 min when a substantial amount of contamination was deposited. This is the worst type of contamination; it essentially ruins the sample and prevents high-resolution work. Unfortunately, this severe carbonaceous deposition can occur in much shorter times, even in a few seconds, if the SEM and the sample are both severely contaminated. After the 10-minute irradiation, an image was taken at 1.27 µm HFW and another at 2.54 µm HFW. The overall area doses were approximately 1.02 mC, 31 nC, and 1.46 nC, respectively. The lesser dose at the 1.27 µm HFW area resulted in only darkening, indicating the second level of contamination. This slight darkening is due to the lower SE yield of the few molecular layers of carbonaceous molecules that have been deposited. The sides of the final 2.54 µm HFW image portion are the third level, with the slightest not showing a visible change. Interestingly, even if large amounts of contamination were deposited, the very center of the highest dose area was not contaminated. Here, the desorption of contamination molecules overwhelmed their adsorption.

The 2.54 µm HFW SE image on the right of Fig. 1 shows the sample after being irradiated at 1.27 µm HFW for 10 min with an overall dose of 0.15 mC. The so-called upper, in-column SE detector acquired all SE images throughout this work. With a clean SEM and a clean sample, no darkening, the evidence of contamination deposition is visible. The center part of the sample that received the highest dose of electrons shows a brighter area. This is due to increased SE yield because the surface's high-dose region of the Si chip was oxidized by electron irradiation in the presence of residual H_2O (Xu et al., 1997; Gurbán et al., 2018). This brightening is the fourth desired level, a good indication of the very high level of cleanliness. If the SEM image of this reference sample is this clean, it can be used to image/measure the sample for many hours without any sign of contamination developing. This is the state of ultra-high cleanliness; see Fig. 2 for the various states of cleanliness and their results.

The blue dots represent water, the grey ones are contamination precursors, and the black ones are already deposited carbonaceous contamination molecules. A thin SiO_2 layer is created during electron irradiation in the presence of water molecules. It is not easily noticed or measured if the irradiation time is short. SEs change the precursor molecules so they no longer have high surface mobility. High-energy electrons make the precursor molecules volatile so they can be pumped out. These electrons also make already deposited carbonaceous contamination molecules volatile, but with much less effectiveness. Fig. 2d shows a much thicker deposited carbonaceous contamination because this SEM uses a beam scanning method with which the electron beam waits for the 60 (50) Hz AC mains signal synchronization on the left and the right of the image frame. Because of this, there the dose is higher.

The deposition of carbonaceous contamination also depends on the size of the area irradiated by electrons, which, at high magnifications, forms a circular, sort-of-volcano shape. Fig. 3 on the left shows the SE image of gold nanoparticles (NPs) on a Si chip (2540 nm HFW). The right image (1060 nm HFW) clearly shows the area bombarded at 423 nm HFW with a 5 keV, 43 pA electron beam for 10 min. This area, where the removal of contamination precursor and deposited contamination molecules overwhelms the deposition, is cleaned. In the presence of water molecules, the irradiated rectangle of the Si substrate got oxidized somewhat; therefore, the SE yield became higher than in other areas of the Si substrate. The Au particles in the irradiated area also got cleaner, their SE yield increased, and at the circular border, they got covered with a layer of carbonaceous contamination. In other areas, they have a small amount of contamination precursor molecules and a thin layer of deposited carbonaceous contamination. In the presence of

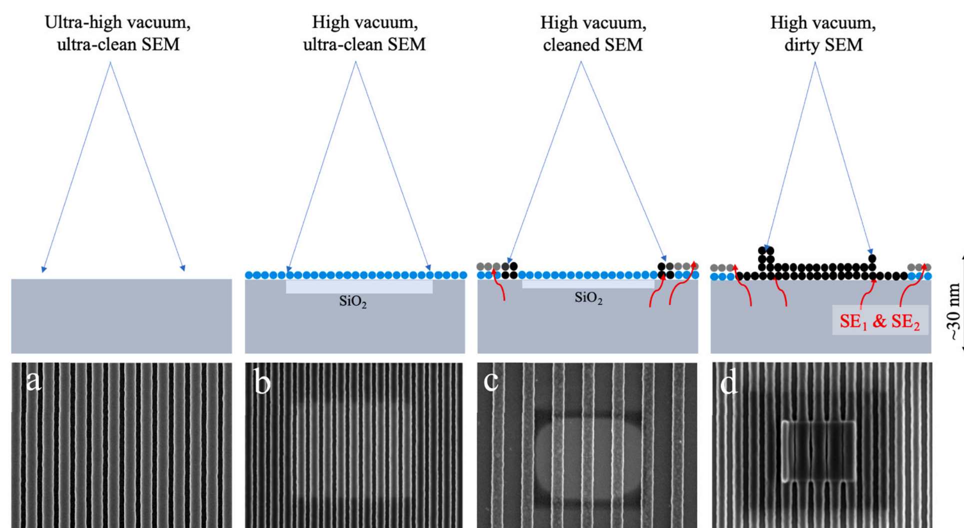

Fig. 2. Contamination deposition depends on the vacuum and the number of precursor molecules. There is no contamination in ultra-high (10^{-7} Pa to 10^{-8} Pa) vacuum (a) and UHC state in high (10^{-3} Pa to 10^{-6} Pa) vacuum (b). Light (c) or severe (d) contamination occurs in unclean high-vacuum systems. HFW is 2.54 µm for all SE 1 keV 43 pA images of the NIST SEMCARS sample.

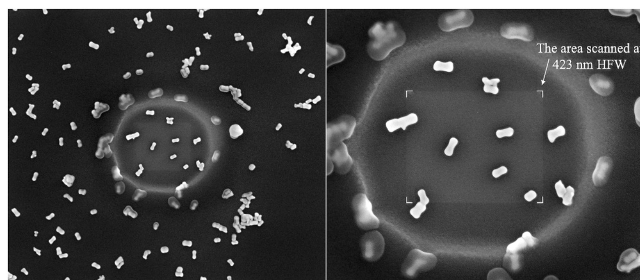

Fig. 3. Deposition of contamination depends on the area irradiated by electrons also. At high magnifications, the deposited contamination forms a circular volcano shape. The SE image of Au NPs on a Si chip, 2540 nm HFW, after it was bombarded with a 5 keV, 43 pA electron beam at 423 nm HFW for 10 min. In-lens SE detector.

water, the precursor molecules have high surface mobility and supply the material that gets deposited in a circular shape after SEs break the molecules' chemical bonds.

2. Experimental results and discussions

Getting rid of carbonaceous contamination is indispensable for high-resolution SEM work. Fortunately, several well-working plasmas, UV light, and *electron*-irradiation-based solutions exist today for cleaning the SEM and its samples.

2.1. Electron irradiation-based cleaning of a vacuum chamber

Electron ionization (EI) has been known for more than a century. It works with electrons that, in high vacuum, interact with solid or gas phase atoms and molecules and produce ions. EI is especially effective on organic compounds with molar masses up to 600 Da and is used in mass spectrometry. The generated ionized fragments can be volatile and pumped away. Electron-stimulated desorption, desorption induced by electronic transitions (DIET), and photochemistry at surfaces have been described, e.g., in (Yates, 2012; Madey and Yates, 1969) Electron irradiation-based sample cleaning has been observed in SEMs, when some but not large amounts of contamination precursor molecules are present (Toth et al., 2009; Mikmeková et al., 2019).

A new and effective cleaning device was used to eliminate precursor molecules and deposited contamination based on low-energy electron irradiation. Fig. 4 shows the programmable control unit and the UHV-compatible 80 mm diameter flood-type electron source cleaning head. The bakeable, UHV-compatible electron source is a heated tungsten helical filament biased at 100 V, so the electron flood it generates comprises about 100 eV energy electrons. This low electron landing energy was the most effective in eliminating contamination precursor molecules and removing already deposited contamination. A 2 mA to 5 mA emission current effectively cleaned the samples and the SEM sample chamber. At four mA, an emission current of about 25 quadrillion (2.5 ×1016) electrons leaves the electron source every second.

The low-energy electron source is a heated filament. Its temperature was measured with a pyrometer in the middle, and it was found to be at about 1900 °C (2173 K) with 12 watts of filament heating power. At this low temperature, tungsten evaporation from the filament and deposition on the sample is negligible. A 100 W 120 V incandescent lightbulb's W filament is heated to about 2500 °C (2773 K) and lasts 1000 h. The conventional SEM's hairpin tungsten cathode is used at 2800 °C (3073 K) to achieve high brightness at a price, up to 100 h of short lifetime. Due to the relatively low-temperature heating of the filament, its operating lifetime in the cleaning system is over 3000 h. A typical electron flood treatment time needed to reach a clean state of the SEM sample chamber is 6–10 h. In the beginning, longer, up to 100 h of

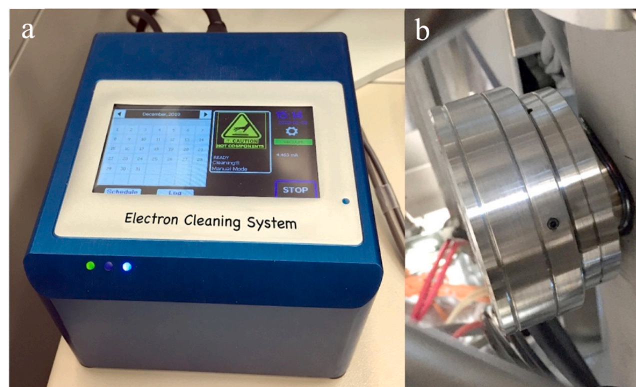

Fig. 4. The programmable control unit of the electron irradiation-based cleaning system (a) and the 80 mm diameter cleaning head (b), the flood-type electron source on the SEM.

cleaning might be necessary to clean a severely contaminated system. It is not unusual to see a recently cleaned system again show contamination, and repeated, shorter cleanings are necessary for contamination-free operation. This is a sign that not all surfaces got cleaned yet because the cleaning only occurs in irradiated areas. Over time, all sources of precursor molecules are exhausted. Once the stable ultra-high cleanliness state has been achieved, only less than 1 h of weekly or by-weekly treatment might be required to keep the SEM contamination-free. Keeping a vacuum chamber open in ambient air for many minutes allows precursor molecules to settle on the surfaces, adding a not negligible source of contamination. The NIST SEMCARS sample and its associated testing procedure are instrumental in checking whether cleaning is necessary.

The electron cleaning device (Fig. 4) has a built-in timer with a selectable cleaning time from 1 to 99 h and a programmable schedule for automatic cleaning cycles.

Fig. 5 shows the residual gas analyzer (RGA) measurement results before and after a weekend-long vacuum chamber cleaning built to study contamination removal techniques and their effectiveness in eliminating precursor molecules and in sample cleaning. The RGA indicated the presence of water, carbon dioxide, hydrogen, nitrogen, and various contamination precursor molecules.

The vacuum chamber was open for over an hour to allow precursor molecules to arrive at the surfaces. After pumping the system from ambient air, the pressure reached $2.4*10^{-8}$ torr ($3.2 *10^{-6}$ Pa). After turning on the electron cleaner, as precursor molecules are made volatile, the pressure goes up, and over time, it gradually goes down. After turning off the electron cleaner, the pressure reached $5.9*10^{-10}$ ($7.9*10^{-8}$ Pa). This was a weekend-long electron irradiation treatment. Some of the vacuum improvement came from pumping the system longer. SEM sample chambers that have already reached their end vacuum have seen a smaller, half to one order of magnitude pressure improvement. The extent of pressure improvement and the time it can be achieved depend on the cleanliness of the SEM sample chamber.

2.2. Electron irradiation-based cleaning of the SEM sample chamber

One advantage of this technique is that it can eliminate contamination whenever it appears. Because it works in a high or ultra-high vacuum, the low-energy electron source can be turned on and starts working instantaneously. For high-quality imaging and measurements, cleaning the SEM to reach the UHC state is indispensable. After that, if necessary, a (few) 10 min of treatment is adequate to return to the UHC state.

Because the electron cleaner uses about 4 mA current of 100 eV electrons, many type 1 and 2 SEs are generated. Preventing these from entering the SEM's electron detectors is necessary. The electron emitter is glowing with a yellowish light, and these photons are also detectable. To avoid oversaturating the detectors, they should be turned off.

2.3. Electron irradiation-based cleaning of the SEM samples

One of the advantages of this technique is that it can be used to clean already deposited contamination whenever it appears. Depending on the cleanliness of the sample, the SEM, and the already deposited contamination, this in-situ cleaning may take longer.

The best practice is to keep the SEM clean, use only clean samples for imaging or measurement, and, if necessary, clean the sample in a separate, dedicated device to avoid the deposition of large amounts of contamination. Using a different electron flood head and a small vacuum chamber for sample cleaning results in much faster cleaning due to the considerably higher low-energy electron dose.

Fig. 6 shows images of nominally 50 nm diameter highly spherical gold nanoparticles deposited on a Si chip. The picture on the left was taken first; it shows that some extra material (likely from the citrate buffer from the colloidal solution) is on and around the Au particles. The image in the middle was taken after 10 min of continuous electron irradiation; the irradiated particles look almost clean. The drift of the sample stage serendipitously allows for the evaluation of the cleaning

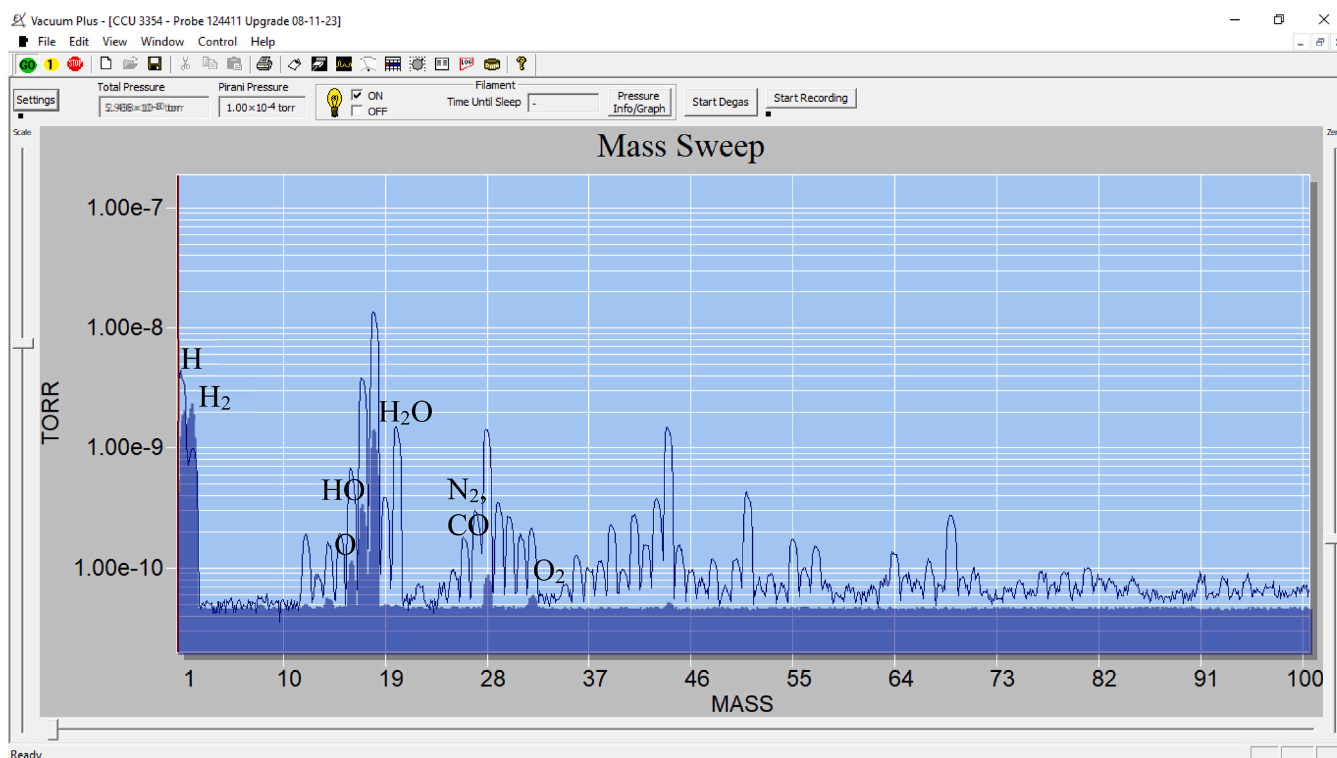

Fig. 5. RGA measurement results were obtained before the electron flood cleaner was turned on (blue line graph) and after a weekend-long cleaning of an unclean vacuum chamber when the electron flood cleaner was turned off (solid blue graph). The vacuum has improved, and many molecular compounds, including hydrocarbon molecules, were removed.

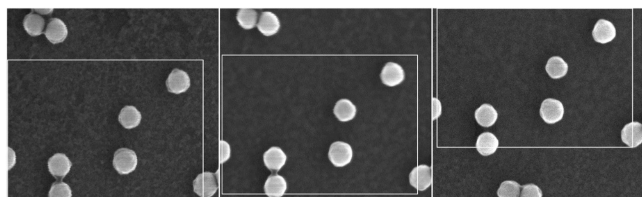

Fig. 6. First, SE image (a) approximately 60 nm diameter Au particles covered with some left-over colloidal material; the image after 10 min of continuous electron irradiation (b), some cleaning is observable; after ten more minutes of irradiation (c), the particles irradiated for 10 min are clean. All 15 keV 80 pA SE images were taken at 512 nm HFW with an in-lens detector.

effect. The particles became clean after ten more minutes of electron irradiation; even the "neck" between two adjacent particles disappeared. Note that the SE yield has also improved due to electron irradiation cleaning. The clean particles are significantly brighter, and the background surface becomes smoother.

The measured beam current by a Faraday cup was 82 pA. The dose over the irradiated areas for 10 minutes was 2.23×10^{-13} C/nm^2, and the total dose over the illuminated regions for 20 min was 4.46×10^{-13} C/nm^2. The Au nanoparticles over the entire sample became clean after the electron cleaner was applied with a similar dose. The UHC state of cleanliness allows for a more accurate size determination with the SEM.

3. Conclusions

Working in a high vacuum with an electron cloud cleaner comes with advantages. The time to and from the actual cleaning is seconds, and most importantly, the contamination removal rate is much higher in a high vacuum than in a lower vacuum. This allows for gentle, low-power cleaning: 2–4 mA electron emission and 100 eV energy electrons result in little energy load on the SEM sample chamber, sample stage, and the sample. Proper care must be taken to avoid overwhelming the SE detectors or removing sample stage lubrication with excessive, unnecessary cleaning.

Another advantage of the system is obtaining a qualitative measurement of contamination in the chamber. This allows the vacuum system to reach a base vacuum level before starting the electron-based cleaning system. Once the base vacuum is reached, the cleaning system is turned on. After the cleaning system has been on for ten minutes, the vacuum pressure is measured and compared with the base vacuum. A change of 1 × 10–4 Pa for typical systems indicates an unclean system that requires approx. 4–8 h to reach complete cleanliness. In a clean system, the change will only be 1×10^{-5} Pa to 2×10^{-5} Pa, so less than 1 h is typically all that is usually needed to reach an ultra-clean chamber.

Low-energy electron irradiation-based cleaning of the SEM sample chamber and/or its samples is an effective technique with advantages over other cleaning techniques used to eliminate precursor molecules of electron-beam-induced contamination. Its fast application and treatment times, small size, and excellent compatibility with SEM practice make it a candidate to be designed for and installed in SEMs to work whenever necessary or regularly to ensure that the SEM always works in ultra-high cleanliness.

CRediT authorship contribution statement

András E. Vladár: Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Resources, Software, Supervision, Validation, Visualization, Writing – original draft, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

References

Bond Dissociation Energies of Organic Molecules, 2003, S.J. Blanksby & G.B. Ellison https://doi.org/10.1021/ar020230d & https://en.wikipedia.org/wiki/Bond-dissociation_energy.
Secondary electron emission of graphene-coated copper, M. Cao, et al., https://doi.org/10.1016/j.diamond.2016.09.019 2017.
D. Fowlkes et al. https://doi.org/10.1021/acsanm.7b00342 2018.
Electron irradiation induced amorphous SiO_2 formation at metal oxide/Si interface at room temperature; electron beam writing on interfaces, S. Gurbán, et al. ⟨https://doi.org/10.1038/s41598-018-20537-4⟩ 2018.
T. Lianga, et al. https://doi.org/10.1116/1.2062428 2005.
T.E. Madey and J.T. Yates Jr. https://doi.org/10.1063/1.1672142 1969.
E.M. Mikmeková, et al. https://doi.org/10.1017/S1431927619003234 2019.
Focused electron beam induced deposition of a periodic transparent nano-optic pattern, A. Perentes, et al. ⟨https://doi.org/10.1016/j.mee.2004.02.079⟩ 2004.
The Scanning Electron Microscope Contamination Assessment Reference Sample (SEMCARS) is available through cooperation with NIST; contact the author at andras@nist.gov.
M. Toth, et al. ⟨https://doi.org/10.1063/1.3187926⟩ 2009.
A.E. Vladár, et al. https://doi.org/10.1117/12.436724 2001.
Enhanced silicon oxide film growth on Si (100) using electron impactJ. Xu, et al. ⟨https://doi.org/10.1063/1.366516⟩ 1997.
J.T. Yates Jr, ⟨https://doi.org/10.1063/1.4746798⟩ 2012.