Secondary electron imaging at gas pressures in excess of 1 kPa

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Environmental scanning electron microscopy (ESEM) enables electron imaging of electrical insulators, vacuum-incompatible materials, and dynamic surface modification processes. For example, at pressures in excess of ~500 Pa, H2O can be condensed in situ enabling characterization of liquids,2 thin films at gas-water interfaces,3 and hydrated biomaterials.4 Electron beam induced deposition and electron beam induced etching are achieved by gases that dissociate to produce reactive species under electron irradiation.5,6 The resulting surface modification processes can be imaged in real time by ESEM.7 Charging caused by electron irradiation of insulators is stabilized by ions produced by electron impact ionization of the gas,8 and the achievable charge control is sufficient for high resolution imaging of materials such as photolithographic masks9 and ultralow-density nanoporous dielectrics.10 However, the usefulness of ESEM is limited by the highest pressure tolerable inside the specimen chamber. In existing systems, hydration requires specimen cooling to just above freezing,4 and gas-mediated surface reaction kinetics are often limited by mass transport of precursor molecules.11

In practice, the maximum pressure employed in ESEM is limited by a decrease in image quality with pressure (at pressures greater than ~500 Pa). Consequently, high pressure imaging necessitates the use of high beam currents, giving rise to low resolution (due to electron optical aberrations)12 and rapid degradation of beam sensitive materials. Here we demonstrate high quality secondary electron (SE) imaging at high pressure (2 kPa), using a relatively low beam current (70 pA). This is achieved by optimizing boundary conditions that govern electron beam scatter in the gas, the energy distribution of low energy electrons traversing the gas, dielectric breakdown of the gas, and electron detector collection efficiency. High pressure ESEM will enable imaging of hydrated materials at close to room temperature (~17 °C), and surface modification processes occurring at high pressures.

Experiments were performed using a FEI Quanta field emission gun ESEM, using the sample-detector geometry shown in Fig. 1. The electron beam enters the high pressure specimen chamber through a conical pressure limiting aperture (PLA).13 Electrons emitted from the sample are amplified in a gas ionization cascade,14 and collected using a stainless steel needle-shaped anode connected to an electrically floating preamplifier. Additional imaging was performed using a solid state backscattered electron (BSE) detector, and a standard off-axis ESEM SE detector15 (SSED) consisting of a large area anode (~2.5 cm2) positioned at ~30 mm away from the beam impact point at the sample surface. H2O was used as the imaging gas. In figure captions, P is the pressure, V0 is the electron beam accelerating voltage, I0 is the beam current, and Vg is the anode bias.

Figure 2(a) shows an image of tin spheres in an amorphous carbon matrix acquired using a beam current of 70 pA at a pressure of 2 kPa, using the needle-shaped anode shown in Fig. 1. SSED images obtained at pressures of 133 and 667 Pa are shown in (b) and (c), respectively. A reference solid state BSE detector image acquired in high vacuum is shown in Fig. 2(d). At low pressures (e.g., 133 Pa), SSED images contain pronounced SE contrast such as that indicated in Fig. 2(b). The contrast corresponds to a thin carbonaceous film that had been deposited on the sample by electron irradiation in a high vacuum environment (~10−7 Pa), using residual hydrocarbons as the deposition precursor molecules.6 However, at elevated pressures, the SE component of the imaging signal is reduced or absent from SSED

FIG. 1. (Color online) Charge coupled device image of the experimental setup. The cone acts as a pressure limiting aperture (PLA) (minimum inner diameter=0.5 mm and sample-PLA gap=1.4 mm). Inset: Left, close-up showing the PLA-sample gap (g) and the anode-beam axis separation (x); and right, electron image of the anode tip.
images, as is illustrated by Fig. 2(c) (SE contrast\textsuperscript{12} is desirable because it typically carries more information than BSE contrast). At pressures in excess of \(\sim 1\) kPa, the SSED efficiency is too low for practical imaging (not shown), necessitating the use of high beam currents that lead to low resolution and rapid damage of beam sensitive materials. Here, these problems are alleviated by the optimized detector geometry shown in Fig. 1, which yields high quality images with pronounced SE contrast at pressures as high as 2 kPa [Fig. 2(a)]. Image quality at such high pressures depends critically on the energy distribution of low energy electrons in the gas cascade,\textsuperscript{12} susceptibility of the gas to dielectric breakdown, detector efficiency, and electron beam scatter in the gas, each of which is discussed below.

We start by considering the mean energy (\(E\)) of SEs traveling to the anode along an arbitrary path \(L\). \(E\) increases due to the energy imparted by the anode potential field (\(V\)), and decreases due to inelastic scattering with gas molecules,

\[
\frac{\partial E}{\partial s} = -q_e \frac{\partial V}{\partial s} - \frac{\partial E_i}{\partial s},
\]

where \(\partial E_i/\partial s\) is the rate of change of energy with distance, \(q_e\) is the charge of an electron, \(\partial V/\partial s\) is the anode electric field, and \(\partial E_i/\partial s\) is the mean energy lost (\(E_i\)) per unit distance traveled in the gas. The first term gives rise to a gas ionization cascade that amplifies the electron imaging signal.\textsuperscript{14} Of the inelastic scattering processes, only ionization makes a significant, direct contribution to the gas cascade.\textsuperscript{14} Figure 3 shows the electron mean free path in \(\text{H}_2\text{O}\) calculated at 0.2 and 2 kPa, using cross sections for ionizing and the dominant nonionizing inelastic processes. The majority of SEs and electrons generated in the gas cascade are emitted with energies below the ionization threshold of the gas. In water vapor, these electrons must be accelerated (by \(\partial V/\partial s\)) to energies in excess of 12.6 eV to enable multiplication through ionization.\textsuperscript{14} During this acceleration, the electrons lose energy (\(\partial E_i/\partial s\)) at a rate determined predominantly by the excitation and vibrational mean free paths shown in Fig. 3. The higher the pressure, the greater the energy loss rate, and the smaller the amount of energy that is eventually used up in ionizing collisions that drive the gas cascade and amplify the imaging signal. At energies in excess of 12.6 eV, the fraction of \(E_i\) that contributes to ionization scales with the ratio of the ionization to the remaining inelastic cross sections, \(\sigma_i/(\sigma_o + \sigma_e)\), shown in Fig. 3. The ratio increases with energy. The higher the gas pressure, the lower the mean electron energy, the smaller the fraction of \(E_i\) used up in ionizing collisions, and the lower the gas gain per unit path length (\(G/L\)). The latter scales with the intensity of the anode electric field (=\(q_e \partial V/\partial s\)), which can be used to increase \(G/L\) up to a maximum limited by dielectric breakdown of the gas. Breakdown depends on the number of molecules ionized by each SE,\textsuperscript{14} and can therefore be prevented by minimizing the electron path length in the gas (\(L\)).

A needle-shaped anode generates a nonuniform electric field that is most intense within a small volume in the vicinity of the anode tip. This field geometry is used to make \(E\) sufficiently high to produce an intense ionization cascade, and \(L\) sufficiently small to prevent breakdown of the gas. In addition, the needle-shaped geometry permits anode placement near the beam impact point at the sample surface, so as to achieve a high SE collection efficiency. Anode placement flexibility is important at high pressures where the sample-PLA distance must be small in order to minimize beam scatter by the gas. The extent of beam scatter is given by the product of pressure and beam path length in the gas.\textsuperscript{16} The latter is minimized by placing the PLA close to the sample surface, as is shown in Fig. 1, where we used a sample-PLA gap of 1.4 mm.

For a given sample-PLA configuration, the geometry of the electric field generated by the anode is a function of anode radius of curvature (\(R\)) and anode coordinates. \(R\) affects gas gain, but at the pressures used here, within the limits imposed by gas breakdown, the effects of small changes in \(R\) on gain can be offset by altering the anode bias and coordinates. Very small radii are undesirable, from a practical viewpoint, because of blunting caused by dielectric breakdown of the gas. Very large radii are unsuitable because the intense field required for efficient amplification must be localized over a small volume. Radii similar to that seen in the inset of Fig. 1 were found to yield good, reproducible
imaging performance. These radii are on the order of the minimum electron inelastic mean free path at 2 kPa.

Anode placement plays a critical role in gas cascade amplification. Figure 4 shows plots of the anode input current $I_a$ (which is proportional to the gas cascade output current) as a function of anode coordinates. At an anode-beam axis separation $x_a$ (shown in Fig. 1) of 1.9 mm, $I_a(z_a)$ contains two local maxima ($z_a$=distance from the anode tip to the pole piece, where $z=0$, as shown in Fig. 1). These are attributed to competing effects of anode position on gas amplification. As $z_a$ is decreased (i.e., the anode shown in Fig. 1 is moved up from $z=9.5$ mm), the gap between the anode tip and the conical PLA decreases. Consequently, the electric field strength along the beam axis decreases, thereby reducing the SE extraction efficiency. However, the field intensity near the anode tip increases, thereby increasing the amplification efficiency per electron collected by the anode.

An increase in $x_a$ (e.g., from 1.9 to 5.9 mm, as shown in Fig. 4) causes a decrease in the electric field strength along the beam axis and in the vicinity of the anode tip. Hence, $I_a(z_a)$ decreases with $x_a$.

Detector efficiency can often be improved by biasing surfaces around the sample. However, in the present case, such biasing generally resulted in a decrease in detector efficiency, as is illustrate by the inset of Fig. 4, showing plots of $I_a$ measured as a function of bias applied to an Al sample, and to the conical PLA shown in Fig. 1. A positive PLA bias causes $I_a$ to decrease due to SE collection by the PLA, and to a decrease in electric field strength in the vicinity of the anode tip. Similarly, a negative bias causes $I_a$ to decrease due primarily to SE recollection by the sample. The curve of $I_a$ versus the sample bias (inset of Fig. 4) shows analogous behavior, but positive and negative biases give rise to SE collection by the sample and the PLA, respectively. Differences between the two curves are caused by the asymmetry in the geometry seen in Fig. 1, created by the needle-shaped anode.

Finally, we note that many ESEM systems use a gaseous SE detector with a ring-shaped anode for SE imaging at pressures in the range of $\sim 150$–$600$ Pa. This configuration is unsuitable for imaging at greater pressures because the electric field generated by such an anode is very inefficient at extracting SEs when the PLA-sample gap is as small as needed for imaging at pressures in excess of $\sim 1$ kPa.

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13Pressure limiting apertures and differential pumping are used to separate the high vacuum electron column from the low vacuum specimen chamber (Ref. 1).


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**FIG. 4.** (Color online) Anode current ($I_a$) measured as a function of anode tip $z$ coordinate ($z_a$), at a number of $x$ coordinates ($x_a$), using electrically grounded Al as the specimen. Inset: $I_a$ measured as a function of PLA and sample bias ($P=2$ kPa, $V_o=15$ kV, $I_a=134$ pA, defocused beam, and $V_a=598$ V).