Journal of Microscopy, Vol. 205, Pt 1 January 2002, pp. 96–105
Received 12 April 2001; accepted 15 August 2001

Consequences of positive ions upon imaging in low vacuum scanning electron microscopy

J. P. CRAVEN, F. S. BAKER, B. L. THIEL & A. M. DONALD
Polymers & Colloids Group, The Cavendish Laboratory, The University of Cambridge Department of Physics, Madingley Road, Cambridge, CB3 0HE, U.K.

Key words. Electron–ion recombination, ESEM, low vacuum SEM, SEM, positive ions, space charge.

Summary

The effects caused by an excess quantity of ionized gas molecules within the low vacuum, variable pressure and environmental scanning electron microscope (ESEM) are described with reference to mechanisms by which they can influence imaging conditions. These effects can include specimen charging, recombination and development of space charge. They are demonstrated for three different classes of sample: (1) an electrically grounded conductor, (2) an electrically floating conductor, and (3) an electrical insulator. A new device is presented that will aid excess charge removal within the ESEM and help correct for some of these effects, thereby dramatically improving imaging over a wide range of operating conditions and samples. The mechanism of image enhancement is demonstrated with reference to the three classes of sample described above.

Introduction

Amplification of the secondary electron signal within low vacuum (LVSEM), variable pressure (VPSEM) and environmental scanning electron microscopes (ESEM) is achieved by multiple cascades of ionizing collisions between electrons emitted from the sample and a low pressure of gas molecules in the chamber (typically 0.5–10 Torr (0.06–1.3 kPa) of water vapour). These cascades are produced by positioning a positively biased electrode, typically 30–600 V, several millimetres above the sample (Danilatos, 1983). In this geometry total amplification gains of up to several thousand times can be achieved in the 'gap' between the sample and the detector (Meredith *et al.*, 1996). One such device that detects the induced signal by utilizing this amplification method is termed the gaseous secondary electron detector (GSED) marketed by the FEI company.

The signal gain obtained from this amplification mechanism is strongly dependent upon the potential difference and distance between the sample and the detector as well as the ionization efficiency and pressure of the environmental gas in the chamber. As the primary beam and backscattered electrons typically have energies greatly in excess of secondary electrons (Goldstein *et al.*, 1992), and the ionization cross-section of water decreases with energy (Paretzke *et al.*, 1986), the gaseous amplification mechanism provides some discrimination between the different signal components (Fletcher *et al.*, 1999). A good discussion of the current understanding of this cascade behaviour is given by Thiel *et al.* (1997a). A direct consequence of the cascade amplification is a residual large concentration of positive ions from the ionized gas. The number of ions formed is equal to the number of electrons produced in the cascade. These ions are repelled by the positive bias on the detector, in the direction of the electric field gradient, towards the sample stage.

Within conventional SEM an insulating sample can quickly build up a large negative potential that rapidly destroys imaging (Cazaux, 1986). Therefore, it is either necessary to image these materials after applying a conductive surface coating to remove excess charge, or to operate the microscope at lower primary beam accelerating voltages, where the net overall specimen potential remains small. This typically occurs when the total electron emission for secondary (δ) and backscattered (η) electrons is greater than unity ($\delta + \eta \geq 1$) (Oatley, 1972). The introduction of a low pressure gaseous atmosphere into a SEM specimen chamber has been observed to reduce charging artefacts dramatically (Moncrieff *et al.*, 1978). When a cascade detection device is used in an atmosphere of a few Torr of water vapour there is a complete absence of the detrimental effects caused by negative specimen charging observed in SEM.

Theoretically, to prevent negative sample charging the flux of positive ions required should not exceed the gain in negative charge from the irradiating electron beam. In an ideal case the number of positive ions formed within the ESEM will exactly

Correspondence: J. P. Craven. Tel.: +44 (0)1223 337007; fax: +44 (0)1223 337000; e-mail: jpc35@phy.cam.ac.uk

balance the negative charge gain. As will be seen in subsequent sections, to achieve sufficient amplification of the electron signal for imaging, these positive ion currents are typically several orders of magnitude greater than the primary beam current and a situation rapidly arises where there is an excess quantity of positive charge in the ESEM chamber. This poses the question whether an excess of positive charge is of concern and detrimental to imaging within the ESEM.

It has recently been recognized that the effects of the positive ion flux in the ESEM may not always be beneficial (Toth & Phillips, 2000) and can be used to explain unusual contrast mechanisms observed under some conditions in the ESEM (Toth *et al.*, 2002).

Space charge and electron–ion recombination

Space charge

The term space charge generically refers to a separation of charged particles in any medium. In the ESEM this term has been used to describe any effects caused by the spatial distribution of electrons and positive ions. The formation of a space charge can become important if the associated electric field is sufficiently intense to modify the detector field experienced by an electron as it traverses the amplification gap. To date there has been very little work exploring the magnitude and effects of a space charge within the ESEM. Within the gaseous amplification process a space charge can occur because ions are more massive than electrons and their mean free path and hence drift velocity in the presence of an electric field are smaller. It is found that the mobility of electrons is three orders of magnitude greater than for the corresponding ions (von Engel, 1955). For water vapour the formation of negative ions due to electron capture of the gas molecules is believed to be insignificant (von Engel, 1955). Therefore, it is only necessary to consider the effects of positive ionized gas molecules and the electrons in the cascade.

The detector–sample configuration within the ESEM is analogous to gaseous amplification between two biased parallel plates. The number of ions formed in the gap increases approximately exponentially towards the anode (detector). Therefore, a large number of ions will have to traverse distances of up to the plate separation gap before they are removed at the cathode (sample). As electrons have a much greater mobility they will reach the anode (detector) on a time scale that will make the ions appear stagnant and each amplification cascade will resemble a 'teardrop' shape (falling towards the detector) with the electrons separated from the ions at the head of the drop. This electrode configuration will exaggerate the space charge effect compared to other geometries, e.g. gaseous amplification between cylindrical electrodes.

In the ESEM it is convenient to refer to a positive ion accumulation in the detector–sample gap as a space charge. The effects of a space charge upon the amplification cascade will depend upon the spatial distribution of ions and location of the 'centre of gravity' of the positive charge accumulation in the volume between the sample and the detector. This will be dependent upon the exact microscope operating conditions, geometry of the detector field and electronic properties of the sample. A more rigorous treatment is beyond the scope of this paper. The modification of the total field experienced by an electron as it traverses the detector sample gap, due to a space charge, has been modelled analytically (Thiel *et al.*, 1997b). In this problem the field produced by ions formed in the gap is integrated over a lateral volume comparable with their diffusion distance. It is concluded that it is not possible for the concentration of positive ions produced in the gap due to ionizing events to collectively produce a sufficient field to distort the detector field and therefore modify the overall amplification characteristics. However, this model assumes that the ions are efficiently removed from the system when they reach a termination point of the detector field (e.g. the sample surface) and there is no temporal accumulation of positive charge. This implies that the recombination electrode for positive ions is a perfect sink. Realistically, recombination rates at sample surfaces are not this efficient and differ over orders of magnitude depending on whether the sample is conducting or insulating. Therefore, if the recombination rate becomes the limiting factor it may still become possible for a significant space charge to accumulate in the detector–sample gap. Experimental observations of the effects of space charges within the ESEM have been presented by Toth & Phillips (2000). They demonstrated how a space charge could become large enough to change the contrast observed whilst imaging an irradiated area in a sapphire insulating sample. The model they propose implies that the efficiency of positive ion removal at the sample surface is a dominant factor in space charge formation.

Electron–ion recombination

Within the ESEM it is necessary to define what is meant by recombination. There are numerous mechanisms whereby an electron can recombine with a positive ion, and it is important to distinguish between those occurring within the ionized gaseous environment (SE–ion recombination, referred to in this paper as scavenging), with those taking place at the surface of a solid material (thermalized and hot e^- ion recombination). (von Engel, 1955; Franzen & Cochran, 1962; Hahn, 1997; Toth *et al.*, 2002).

A conductive surface will act as a source of electrons for incident positive ions providing an efficient mechanism for surface electron–ion recombination. It is this neutralization mechanism that is likely to be dominant, occurring at any conductive surface at which positive ions are incident. These will normally be positions where the detector field terminates e.g. the sample, sample stub and microscope stage.

A poor surface recombination efficiency may result in the formation of a space charge as described above. However, the direct effects of surface recombination upon imaging within the ESEM become of interest if it actually occurs on the sample itself. In this case it is necessary to consider any consequences, due to a flux of positive ions being neutralized at the sample upon the simultaneous electron emission from that region. It is known that a positive ion situated on the surface of a conductor will modify the escape barrier for an electron leaving the sample. The effect that this will have upon the distribution of secondary electron energies leaving the sample and hence image formation in the ESEM is the subject of current research (Toth *et al.*, 2002).

Electron–ion recombination mechanisms in the gas become very important if they result in a loss of the electron-imaging signal. This type of recombination will act as a 'scavenging contribution'. Previous opinion (Danilatos, 1983) has concluded that, under typical ESEM conditions of low pressure and high fields, the ions move too slowly relative to the electrons and do not exist in sufficient concentrations for the probability of an electron being captured by an ion to become significant. It was concluded that the portion of the electron signal lost by this process is likely to be very small. These conditions, however, are unlikely to apply in a small volume just above the sample surface, where the flux of ions arriving at the sample may become greater than the rate at which they are removed by surface electron–ion recombination. Under these conditions an accumulation of positive ions, which may not be sufficient to produce a significant modification of the detector field, may yet be sufficient to scavenge a significant portion of the SE signal. The kinetic energy of emitted secondary electrons from a metal is typically very small (with a Maxwellian energy distribution centred on ~2–3 eV) (Dekker, 1955). Once these leave the sample surface they are quickly accelerated by the detector field and their kinetic energy increases correspondingly. However, before these electrons gain kinetic energy they must (for reasons stated above) traverse a region of high ion concentration. Within this region it is proposed that the probability of capture for a secondary electron by a positive ion is greatly enhanced and therefore the total gaseous electron–ion recombination is significantly increased. It is also worth noting that within this near surface volume, before significant gaseous amplification has occurred, the loss of a few electrons due to scavenging is likely to result in a dramatic decrease in the total imaging signal.

Classification of materials

Not all materials will have the same response to the presence of an electric field and irradiation by charged particles. Therefore, to simplify explanations and observed effects for this paper, all samples have been generalized into three different classes of material, each with a different response. These classes are: (1) an electrically grounded conductor, (2) an electrically floating (isolated) conductor and (3) an electrical insulator.

A general overview of the effects encountered during imaging of each of these samples in the ESEM is discussed in the subsequent section.

Grounded conductor

The simplest material to consider is an electrically grounded conductor. It is not possible for this sample to sustain any internal electrical field due to the delocalization of charge carriers over the sample surface. Therefore, any external fields, e.g. the detector field, must terminate on the sample surface. The electrical contact with earth through the sample stage ensures that any imbalance of charge, caused either by electron beam or ion irradiation, arriving at the sample, will be quickly compensated by a flow of electrons to or from ground, respectively. Under these conditions the detector field strength between the sample and the detector will remain constant, ensuring consistent amplification. As charge carriers in the gap between the sample and the detector are quickly removed, then it is unlikely that a significant space charge can be sustained in the gap to distort the detector field enough to change the amplification characteristics. Thus, as is frequently observed in practice, few problems are encountered when imaging these samples in the ESEM. However, under conditions of high gas pressure and detector bias, where a large quantity of ions are produced in the amplification cascade, the flux of ions towards the sample surface will be large and then it is necessary to consider whether effects of SE–ion recombination are significant.

Floating conductor

The situation with a floating conductor is very similar to the case of the grounded conductor described above. As before, the sample is not capable of sustaining an internal field because charge carriers are free to redistribute within the material. However, in this case there is no external circuit to earth to remove any charge imbalance. If this material is placed in the gap between the sample stage and the detector it will assume a net potential, induced by its position in the electric field. Providing the sample-stage is held at earth then this potential will be between 0 and 600 V depending upon its position relative to the detector. Because the sample will have a finite volume and must assume the same potential over its entire surface then this will lead to a modification of the detector field. As the sample is above ground potential then a second field must also exist between the bottom surface of the sample and the grounded sample stage. In practice, the volume occupied by this field will be filled with a dielectric medium to electrically isolate the sample from the stage. This will reduce the field strength within this region and therefore lower the assumed potential of the floating conductor. When charge carriers arrive at the surface of the sample, either from the electron beam or the positive ions from the gaseous amplification, then the potential will change according to the balance of each species of charge carriers arriving at the plate. If there is an excess of positive ions then the sample will quickly assume a positive potential. The assumed potential will then be due to the degree of charge imbalance and the capacitance of the arrangement.

As the potential of the sample becomes more positive then the field strength between the sample and the detector will be smaller and hence the total amplification reduced. As the amplification decays, then the ion current reaching the sample surface will reduce and after a period a dynamic equilibrium will be reached where the balance of positive and negative charge carriers reaching the sample will be equal and the potential of the sample becomes constant. The change in potential of a floating conductor can be easily demonstrated experimentally in the ESEM by measuring the potential with respect to ground of the sample with a high impedance electrometer. Figure 1 shows the increase in sample potential as a function of gas pressure (hence ion flux) of a conducting copper sample ~2 cm × 2 cm electrically isolated from the grounded sample stage by a thin sheet of latex. It can be seen that as the gas pressure is increased from 0.1 Torr (0.013 kPa) the plate potential quickly becomes positive at ~0.2 Torr (0.026 kPa) and increases up to a maximum of ~120 V at 2 Torr (0.26 kPa). This corresponds to a reduction in the original detector field (detector bias 300 V) by almost a factor of 2. It is also interesting to observe that the conditions for charge neutrality occur at an extremely low gas pressure (~0.2 Torr, 0.026 kPa) at this detector bias. When using a GSED detector, typical operating conditions to ensure sufficient amplification are between 1.5 and 2 Torr (0.2 and 0.26 kPa) of water vapour with a detector bias of ~300–500 V. This demonstrates that under most conditions the sample will be predominantly exposed to an excess flux of positive ions. It is worth noting that the increase in sample potential will not inhibit electron emission by 'pinning' electrons to the surface. This is because the result of the positive potential is that the entire sample assumes a higher potential energy, but there is still a net field to the detector.

Obviously the effects of a reduced detector field are detrimental for imaging the sample surface. However, as in the case of a grounded conductor, it is still necessary to consider any possible effects of recombination. In this case, the effects should be the same for a grounded conductor under conditions where the detector field is equivalent to the residual field on the floating sample.

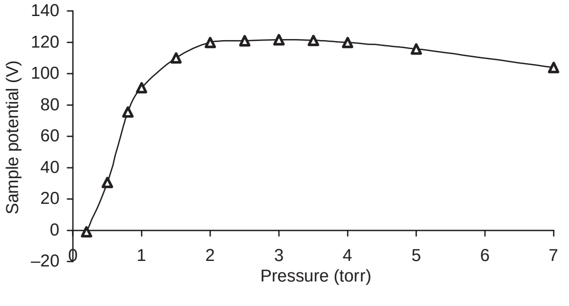

Fig. 1. The change in sample potential for a copper plate imaged in the ESEM as a function of gas pressure. Detector bias = 300 V, working distance = 9 mm.

Insulator

In the case of an insulating material the situation becomes a great deal more complex. Charge within insulators is not free to redistribute within the material and therefore they have the ability to sustain an internal field. When the sample is placed in the ESEM, the detector field will not terminate on the sample surface, as in the previous two cases, but will penetrate through the insulator and terminate on the grounded sample stage. The field strength within the sample will be determined by the dielectric constant of the sample and generally be reduced from that in the vacuum above the sample. Upon irradiation with an electron beam, further complex fields are set up within the interaction volume of the sample due to the implantation and loss of charge from different depths. A generalized analysis of a typical arrangement is difficult because the result is dependent upon the exact properties of the sample and incident irradiating beam. A detailed description, dealing with several different arrangements is given by Cazaux (1999). Within the ESEM the situation is further complicated by the addition of the positive ions from the amplification cascade. These will follow the detector field from their point of creation and accumulate at the sample surface. Any assembly will have the effect of providing the sample with a surface coating of positive charge. In small quantities these ions will be preferentially attracted to any areas of negative surface potential on the sample surface and compensate for detrimental imaging artefacts seen when putting these materials in conventional SEMs. In larger concentration a coating of positive charge may have numerous effects including: an accumulation of a positive space charge that will distort the detector field (Toth & Phillips, 2000); creating a medium in which the probability of gaseous recombination of electrons leaving the sample is increased and imaging signal is scavenged (Toth *et al.*, 2002), and a modification of the escape barrier for electron emission from the sample surface.

Field modification device

The device

It has been established for many years in SEM that the use of conducting paths to ground in the form of grounded stubs, foil containers and conductive paints and tapes that are very close to the point of imaging helps alleviate charging on problematical samples (Pfefferkorn *et al.*, 1972). Indeed, this approach can also be used within the ESEM to establish an effective ground plane at the sample surface for the detector field

(Newbury, 1996). Typical methods, however, are often very restrictive as they limit the area of observation and cannot be used universally for all samples, especially when performing dynamic hydration experiments and imaging 'wet' samples. In the current study a device (Craven & Baker, 2000) was constructed consisting of a parallel array of fine gauge (< 100 μm diameter) conductive wires spaced at intervals of ~2 mm as shown schematically in Fig. 2. This device was mounted in the gap between the sample and the detector, supported independently of the sample stage and positioned so there was no electrical contact between the sample and the device. The spacing between the wires and the sample surface was typically of the order of ~1–2 mm. The wires on the device were all electrically connected to earth to ensure they maintained a constant 0 V potential.

Device effects

Inserting a conductor into the detector field in this manner will have two main effects. Firstly it will introduce new regions into the gap that are maintained at earth potential that will act as termination points for the detector field above the sample. Secondly, a large proportion of positive ions will follow the detector field to these new termination points and the device will act as a new recombination centre for positive charge. The effect of the device on the detector field is shown schematically in Fig. 3.

There are several points worth noting about the design. The wires must be spaced so that the detector field is not completely screened from the sample surface and there is still some penetration through the device to the sample surface. If the spacing of the wires is too small then the device will screen the sample from the detector field. In this case the field gradient will be too small to accelerate secondary electrons efficiently towards the detector, and only secondary electrons that diffuse across the plane of the grid will be amplified and detected. The perturbation of the detector field introduced by the device has been modelled by solving Laplace's equation in 2-D for one grid spacing of the device mounted between two infinitely long parallel plate electrodes (Fig. 4). In this case the device wires were 100 μm in diameter, spaced at 2 mm intervals, and the device was placed 1.5 mm from the sample surface in a total detector–sample gap of 5 mm. From this figure it is possible to see that between the wires there is still penetration of the detector field to the sample surface.

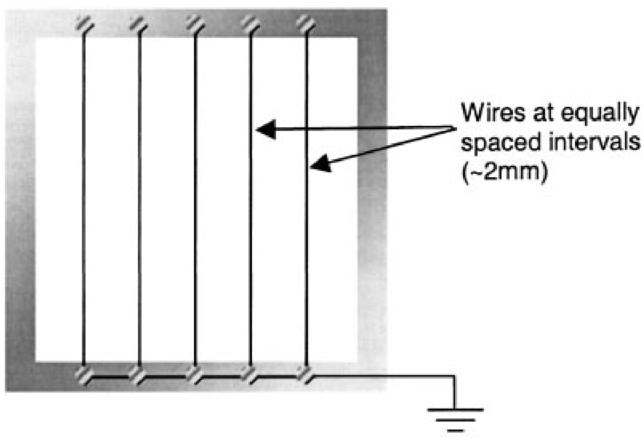

Fig. 2. A schematic representation of the device showing a series of parallel wires (approximate separation = 2 mm) electrically connected to ground.

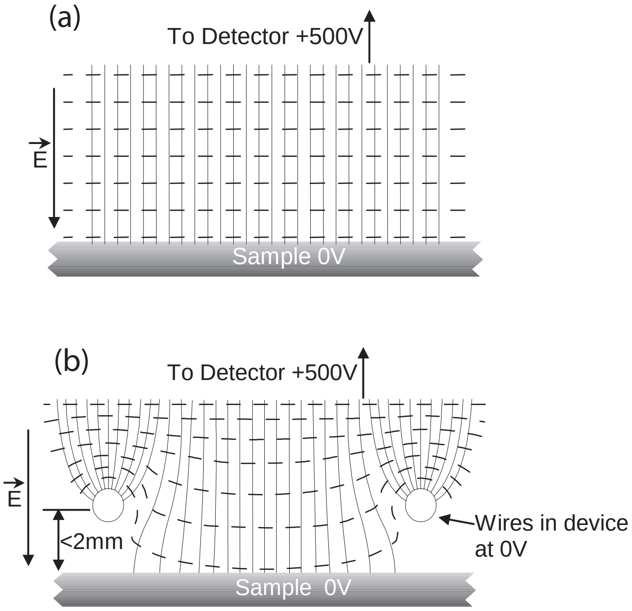

Fig. 3. A schematic of the typical electric field achieved in the ESEM, between the detector and grounded conductive sample assuming a parallel plate configuration, without (a) and with (b) the device in position. Only the fraction of the field above the sample is shown in (a) and (b). In (b) device wires are drawn with long axis normal to plane of page. Only two device wires are drawn for clarity. Solid lines represent field lines and broken lines represent lines of equipotential.

By keeping the diameter of the wires on the device very small, it is also possible to intensify the field gradient around the wires. This should have the beneficial effect of accelerating the positive ions to these points so they can be removed more efficiently. This device was inserted above examples of the three classes of materials outlined previously. Experiments were conducted either in an Electroscan Model E3 ESEM equipped with a LaB_6 filament or an Electroscan Model 2010 ESEM equipped with a tungsten hairpin filament. The following section describes the effects of imaging with the device in place, with practical examples of each case.

Results

Grounded conductor

Figure 5 shows the effect of the device on a grounded conducting plate. In this case the sample was a copper plate (surface

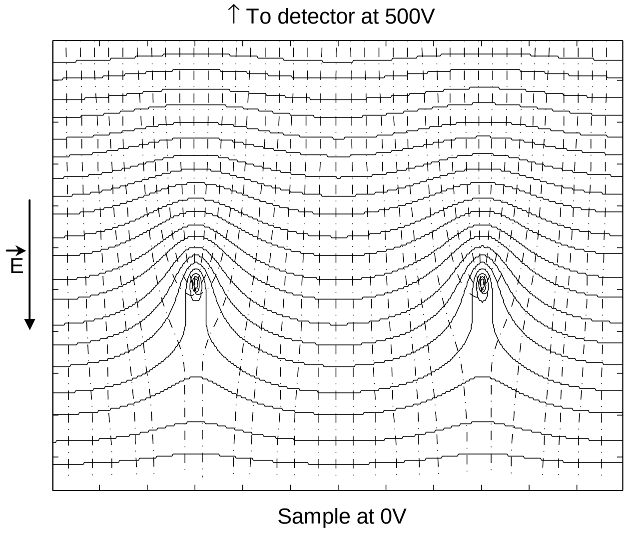

Fig. 4. Computer calculation of the detector field with the device in place (assuming that the detector and sample can be modelled as two infinitely long parallel plates). Only the fraction of the field above the sample has been plotted. Solid lines represent lines of equipotential. Broken lines represent the detector field. Device mounted 2 mm from sample, wires spaced at 2 mm intervals with detector sample spacing = 5 mm. Relative wire diameter = 100 µm. Detector bias = 500 V.

area ~2 cm × 2 cm). An overall increase in the signal is obvious when the device is inserted, as shown in Fig. 5(b). In fact, the brightness control (i.e. dc offset) had to be reduced in Fig. 5(b) in order not to saturate the image! If the beam is placed into 'spot mode', the signal intensity, collected by the GSED, can be measured as a voltage output from the detector pre-amplifier. Figure 6 shows the variation in signal as a function of gas pressure with and without the device in place. As the working distance to the sample surface was kept constant in both cases, it might be assumed incorrectly that inserting the device into the gap between the sample and the surface modifies the detector field and affects the amplification. Following the theoretical model of Thiel *et al.* (1997a), increasing the detector field by decreasing the gap size will shift the maximum amplification peak towards higher pressures. Figure 7 demonstrates that at pressures below the amplification maximum (i) (curve A) inserting the device should actually decrease the amplification if it was indeed a field effect. Conversely at higher pressures (ii), inserting the device should increase the amplification. Again, Fig. 6 illustrates that the signal is increased throughout the pressure range used. Furthermore, no lateral shift in the amplification peak is evident in Fig. 6. As has been argued earlier, space charge effects will be negligible above a grounded conductor, and as specimen charging is impossible, the only remaining explanation for the signal increase is that the device has allowed more electrons to enter the amplification cascade, suggesting that electron–ion recombination mechanisms have been reduced.

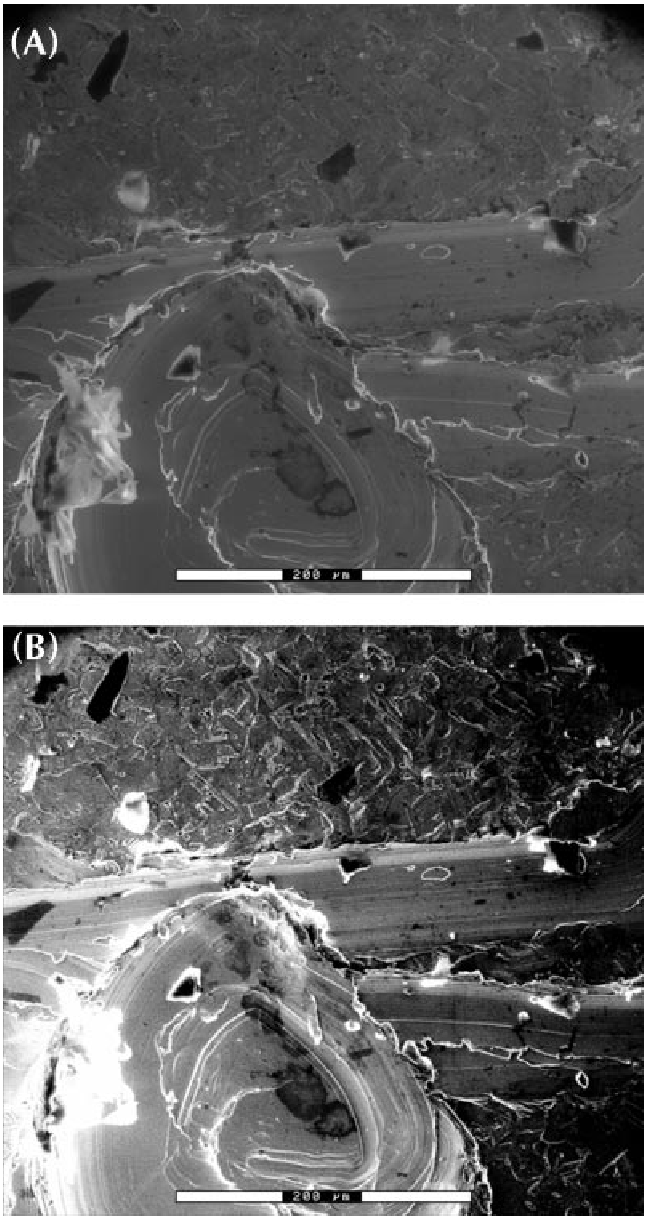

Fig. 5. (a) ESEM image of a grounded copper plate without the device in position. (Beam energy = 20 keV, gas pressure = 3.5 Torr (0.45 kPa), working distance = 8.7 mm). (b) As (a), but with device in position. The dc offset of the GSED pre-amplifier has been reduced to reduce detector saturation. Device wires at 1 mm from sample surface. Note the signal increase around asperities and edge regions.

On further inspection of Fig. 5(b) it is apparent that much of the enhanced signal originates from edges of asperities on surface features. This observation suggests a disproportionate increase in the electron signal from these areas. Without the device in place, these localized features are likely to introduce small distortions into the electric field, concentrating the field gradient in these areas. This will cause these regions to preferentially act as recombination sites for positive ions, and

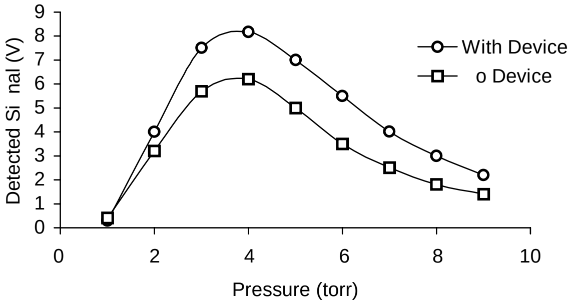

Fig. 6. Total GSED signal measured from a grounded copper sample for a detector–sample gap size of 5 mm, with and without device in place (detector bias = 450 V). Note the change in the height of the amplification peak; however, there is no lateral shift in the peak position with pressure. Device wires 1 mm from sample surface.

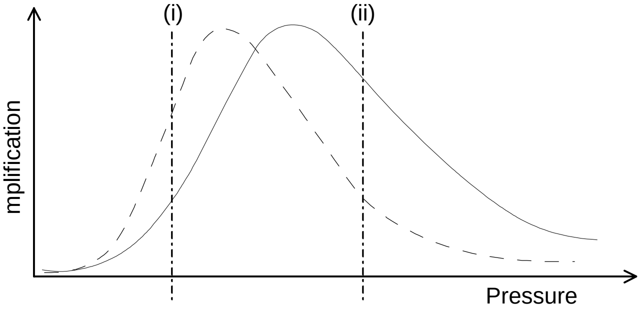

Fig. 7. A schematic representation of the change in theoretical total signal amplification with change in gap size at constant detector bias as a function of gas pressure. Gap size for curve A is smaller than for curve B.

therefore any gaseous recombination of electrons leaving the sample may be more significant in these areas (Toth *et al.*, 2002). This effect is decreased when the device is inserted into the gap because the detector field is modified so that the ions will tend to recombine on the device wires.

Floating conductor

A conducting copper plate was electrically isolated from the sample stage and allowed to equilibrate to a floating potential. Figure 8 displays the effect of imaging the copper plate with and without the device in place. It is obvious that there is a significant enhancement in contrast in Fig. 8(b) compared with Fig. 8(a). The effects of the device in this case are more obvious. As shown above (Fig. 1), a floating conductive plate can assume a non-zero potential. This will act to reduce the potential difference and hence the signal amplification between the sample and the detector. By inserting the device in the gap, the current of positive ions arriving at the sample surface and its equipotential are both reduced. Figure 9 shows the results of a simple experiment measuring the sample potential as a function of detector bias. It is clear that there is a large reduction in the potential of the sample when the device is in place. It is worth noting that the potential on the sample still remains substantially positive. This is because there is still a penetration of the detector field through the device and, consequently a (reduced) flux of ions arriving at the sample surface. Therefore, the sample will reach a new potential equilibrium that will not necessarily be at a plane of zero equipotential (Fig. 9). If the sample is at a positive potential below the device (held at earth potential) then a portion of the detector field, between the sample and the device, may be inverted and cause the

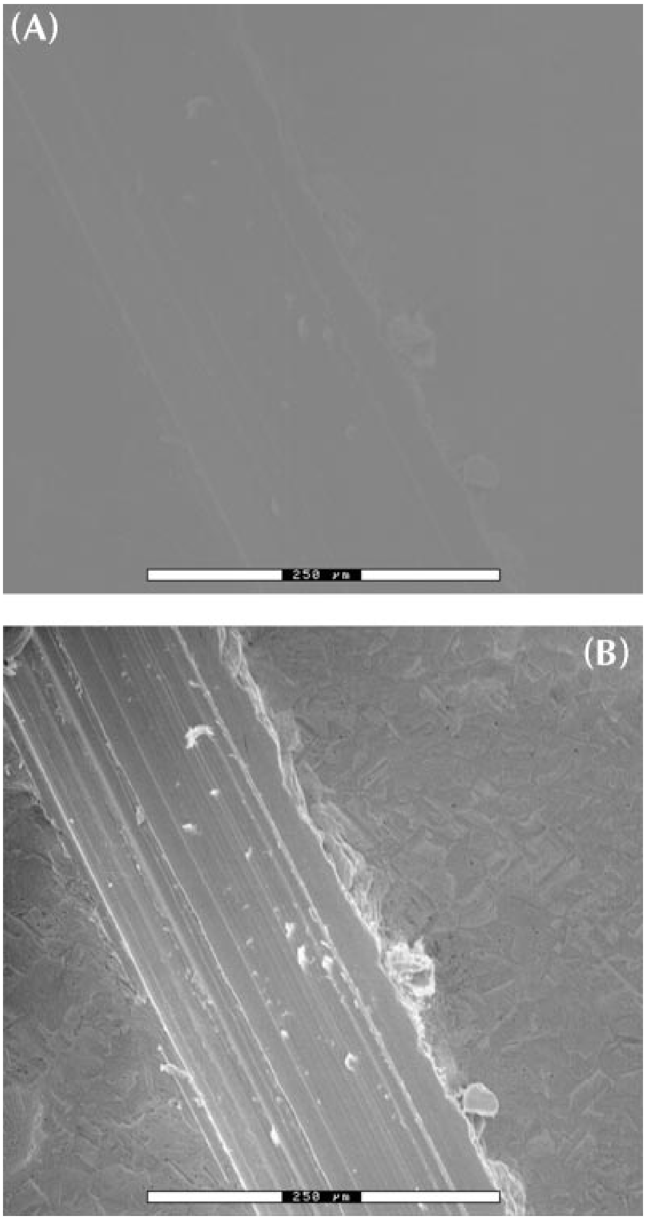

Fig. 8. (a) ESEM image of a floating copper plate without the device in position. (beam energy = 20 keV, gas pressure = 2.9 Torr (0.38 kPa), working distance = 13.8 mm). (b) As (a), but with device in position. Device wires 1 mm from sample surface. Note the restoration of image contrast.

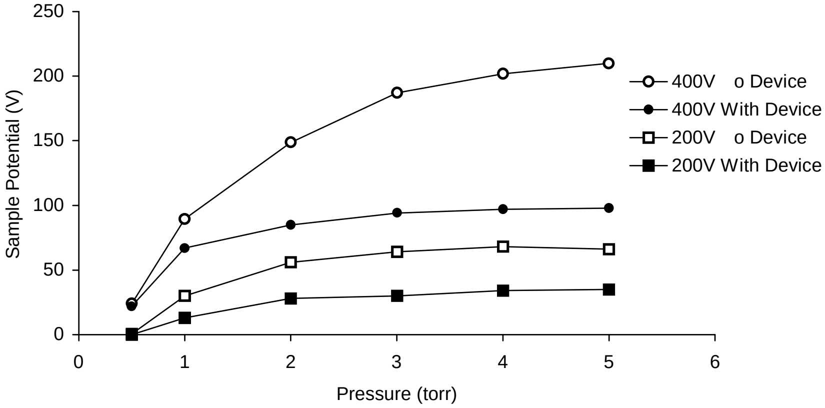

Fig. 9. Measured sample potential curves as a function of gas pressure for a floating copper plate (Fig. 8) with and without the device in position for two different detector biases. (Beam energy = 20 keV, working distance = 13.8 mm).

secondary electrons to be 'pinned' at the sample surface. This will decrease the secondary electron content in the image and therefore it is likely that Fig. 8(b) contains a higher proportion of backscattered signal information. Therefore, although the device will have the effect of lowering the potential of the sample and restoring a sufficient detector field for imaging conditions (see also Figs 8(a) and (b)), there still remains a problem if it is required to know the exact signal composition.

Insulator

Figures 10(a) and (b) show an insulating polymeric material on the surface of a quartz slide. Both images are obtained under identical experimental conditions except for the introduction of the device in Fig. 10(b). The image histogram has been expanded to enhance the contrast in Fig. 10(a) so that the image is visible. The difference in image quality is very dramatic. From Fig. 10(a) it is possible to see a dark band around the polymer at the interface with the quartz slide. This is indicative of a differential charging between the two materials establishing a resultant field and inhibiting electron emission from the quartz plate around the polymer. In Fig. 10(b) this effect is not so noticeable and the presence of fine structure in the quartz plate is also visible. Similar to the case of a floating conductor, this improvement in image quality is an indication of an increased amplification field introduced by inserting the device in the gap. Again, the device will also reduce the flux of positive ions arriving at the sample surface and aid the removal of any space charge accumulation above the sample surface.

It may be expected that removing the flux of positive ions arriving at the sample surface will reintroduce the charging problems observed when imaging these materials in the

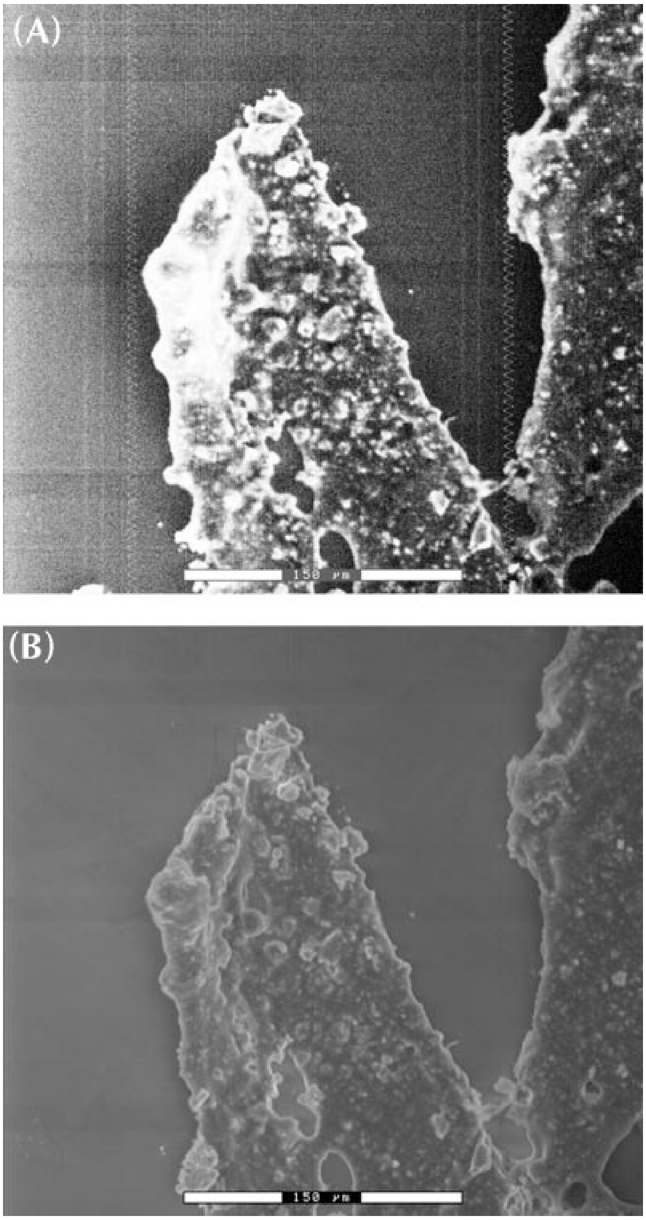

Fig. 10. (a) ESEM image of a polymer on a quartz plate without the device in position. Image histogram has been stretched to show contrast (beam energy = 20 keV, gas pressure = 2.9 Torr (0.38 kPa), working distance = 12 mm). (b) As (a), but with device in position. Device wires 1 mm from sample surface.

absence of a gaseous environment. In practice this is not observed, provided that there are sufficient ions in the chamber, as the device will act via a self-compensating mechanism. If part of the sample surface assumes a negative potential, the detector field will be modified so there will be a preferential flow of ions to the sample surface instead of the device.

There has been much interest regarding the imaging of insulating materials under conditions in the ESEM, where additional contrast due to electronic defects is observed in the ESEM (typically less than 1 Torr (0.13 kPa) of water vapour) (Schnarr & Futing, 1997; Griffin, 2000). This contrast has been termed 'charge contrast imaging' (CCI) and although observed clearly at low pressures, contrast is quickly lost as pressure is increased. The ESEM conditions under which CCI is observed correspond roughly to those of charge neutrality where there are only just enough ions produced in the cascade to prevent negative charging. Under these conditions, local changes in surface potential are likely to arise due to regions of different trapped charge density giving rise to a form of voltage contrast (Cazaux, 1999). As the pressure of gas and detector bias are increased, and correspondingly the positive ion flux, this contrast is rapidly lost. Therefore, it is apparent that an excess quantity of positive charge is detrimental for this contrast mechanism. With the device in position the range of ESEM conditions over which CCI has been observed is greatly extended and has been clearly observed over the entire pressure range of the machine where normally this would not be possible. This is exemplified by Figs 11(a) and (b), which were acquired at a pressure of 4 Torr (0.52 kPa). In Fig. 11(b) with the device in position, 'charge contrast' is very apparent. When the device is removed (Fig. 11(a)) the 'charge contrast' completely disappears. This extension of imaging conditions over which CCI imaging is observed is consistent with the device removing excess positive charge and restoring the detector field above the sample.

Conclusions

The effects of an excess quantity of positive ions can cause severe degradation of secondary electron imaging within the ESEM. The mechanism by which this occurs is sample-dependent, and the most probable explanations have been outlined for three different classes of material. A simple field restoration/ion removal device has been constructed that improves imaging for a variety of samples and conditions. In each class of material the improvement is consistent with the introduction of new electron–ion recombination regions for positive ions and the restoration of a sufficient potential gradient between the sample and the detector to achieve an effective gaseous amplification. Although the presence of an enhancement effect above a grounded conductor suggests that a large proportion of electron signal is removed due to gaseous capture and recombination, it is not possible to discount (although it is unlikely to occur above a conductor) the possibility of space charge effects. However, the presence of the device and the subsequent removal of excess positive ions should diminish, if not completely remove these phenomena.

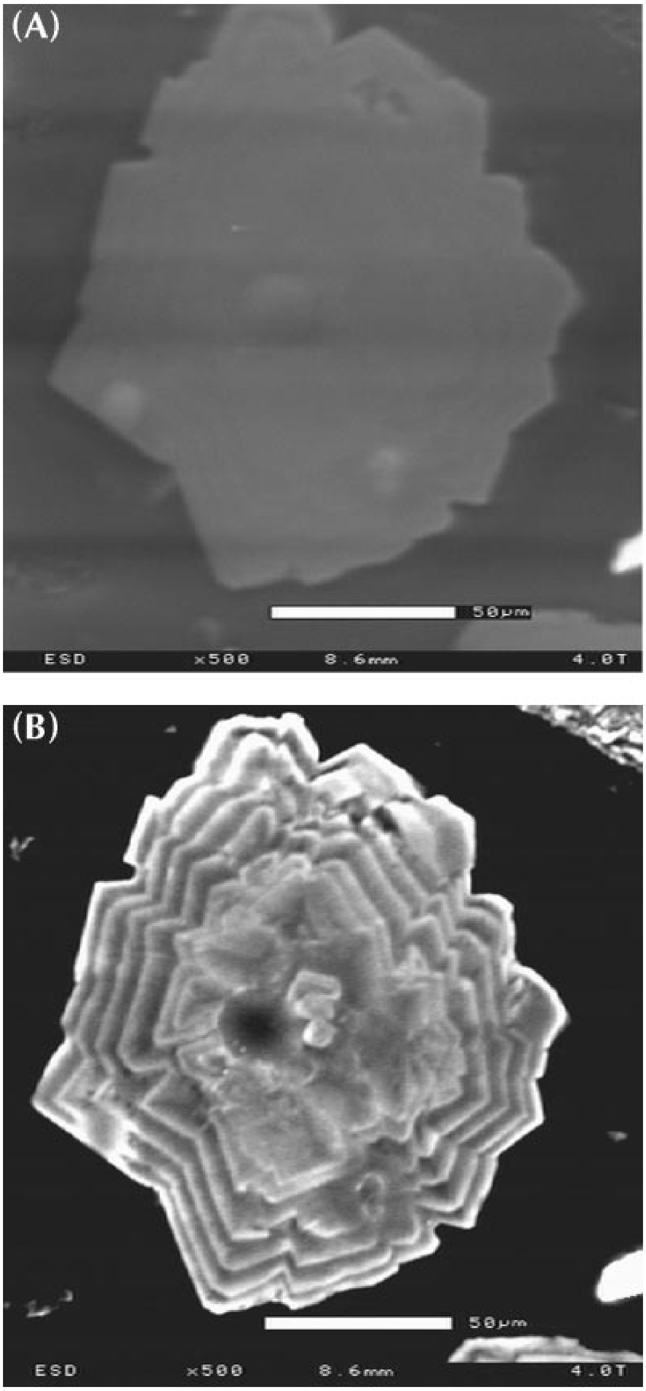

Fig. 11. (a) ESEM image of polished Gibbsite ($Al(OH)_3$) without the device in position. (Beam energy = 20 keV, gas pressure = 4 Torr (0.52 kPa), working distance = 8.6 mm, scan speed = 2.1 frames s^{-1} with eight frame integration). (b) As (a), but with device in position. Device wires 1 mm from sample surface. Note the visibility of 'CCI' contrast.

Acknowledgements

The authors thank Milos Toth for useful discussions and Brendan Griffin for kindly providing the Gibbsite sample. This work was funded by the EPSRC and Cambridge Schlumberger research through a CASE studentship to J.P.C. and the FEI Company (B.L.T.).

References

Cazaux, J. (1986) Some considerations on the electric field induced in insulators by electron bombardment. *J. Appl. Phys*. **59**, 1418–1430.

Cazaux, J. (1999) Some considerations on the secondary electron emission, δ, from e^- irradiated insulators. *J. Appl. Phys*. **85**, 1137–1147.

Craven, J.P. & Baker, F.S. (2000) Patent Application No. 0005717.4. Cambridge University Technical Services Ltd, Cambridge.

Danilatos, G.D. (1983) A gaseous detector device for an environmental SEM. *Micron Microscopica Acta*, **14**, 307–318.

Dekker, A.J. (1955) Secondary electron emission. *Solid State Physics, Vol*. 6 (ed. by F. Seitz and D. Turnbull), pp. 251–311. Academic Press, New York.

von Engel, A. (1955) *Ionized Gases*. Clarendon Press, Oxford.

Fletcher, A.L., Thiel, B.L. & Donald, A.M. (1999) Signal components in the environmental scanning electron microscope. *J. Microsc*. **196**, 26–34.

Franzen, W. & Cochran, L.W. (1962) Pulse ionization chambers and proportional counters. *Nuclear Instruments and Their Uses, Vol. 1* (ed. by A. H. Snell), pp. 3–81. J. Wiley & Sons, New York.

Goldstein, J.I., Newbury, D.E., Echlin, P., Joy, D.C., Romig, A.D., Lyman, C.E., Fiori, C. & Lifshin, E. (1992) *Scanning Electron Microscopy and X–Ray Microanalysis*. Plenum Press, New York.

Griffin, B.J. (2000) Charge contrast imaging of material growth and defects in environmental scanning electron microscopy – linking electron emission and cathodoluminescence. *Scanning*, **22**, 234–242.

Hahn, Y. (1997) Electron-ion recombination processes – an overview. *Report Prog. Phys*. **60**, 691–759.

Meredith, P., Donald, A.M. & Thiel, B.L. (1996) Electron–gas interactions in the environmental SEM's gaseous detector. *Scanning*, **18**, 467–473.

Moncrieff, D.A., Robinson, V.N.E. & Harris, L.B. (1978) Charge neutralisation of insulating surfaces in the SEM by gas ionisation. *J. Phys. D: Appl. Phys*. **11**, 2315–2325.

Newbury, D.E. (1996) Imaging deep holes in structures with gaseous secondary electron detection in the environmental scanning electron microscope. *Scanning*, **18**, 474–482.

Oatley, C.W. (1972) *The Scanning Electron Microscope, Part 1, the Instrument*. Cambridge University Press, Cambridge.

Paretzke, H.G., Turner, J.E., Hamm, R.N., Wright, H.A. & Ritchie, R.H. (1986) Calculated yields and fluctuations for electron degradation in liquid water and water vapour. *J. Chem. Phys*. **84**, 3182–3188.

Pfefferkorn, G.E., Gruter, H. & Pfautsch, M. (1972) Observations on the prevention of specimen charging. *5th Annual Scanning Electron Microscope Symposium, Chicago, Illinois, U.S.A*. IIT Research Institute, Chicago.

Schnarr, H. & Futing, W. (1997) Some aspects of optimizing contrast for the investigation of joint materials in the environmental scanning electron microscope. *Scanning*, **19**, 79–84.

Thiel, B.L., Bache, I.C., Fletcher, A.L., Meredith, P. & Donald, A.M. (1997a) An improved model for gaseous amplification in the environmental SEM. *J. Microsc*. **187**, 143–157.

Thiel, B.L., Hussein-Ismail, M.R. & Donald, A.M. (1997b) *Effects of Space Charge on ESEM Gas Amplification*. Microscopy and Microanalysis. Springer, Cleveland, Ohio.

Toth, M. & Phillips, M.R. (2000) The effects of space charge on contrast in images obtained using the environmental scanning electron microscope. *Scanning*, **22**, 319–325.

Toth, M., Thiel, B.L. & Donald, A.M. (2002) On the role of electron-ion recombination in low vacuum SEM. *J. Microsc*. **205**, 86–95.