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THE BEAM-GAS AND SIGNAL-GAS INTERACTIONS IN THE VARIABLE PRESSURE SCANNING ELECTRON MICROSCOPE

C. Mathieu*

Université d'Artois, Faculté Jean Perrin, Lens Cedex, France

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Abstract

The variable pressure scanning electron microscope (VPSEM) is a promising new family of techniques which permits the imaging of insulators without preparation (coating). The elimination of specimen charging is accomplished in the VPSEM by the introduction of air inside the specimen chamber. Unfortunately, directing an electron beam into a gas creates various interactions between the gas, the beam, the signal, and the specimen. The aims of this study were to correlate probe current measurement, Monte Carlo simulation and X-ray microanalysis is order to optimize the use of the VPSEM. A method has been proposed to determine the scattered cross section of the air gas. The experimental conditions to optimize imaging and X-ray microanalysis are quite similar. However, microanalysis of the light element such as carbon, and oxygen is problematic because the conditions are not optimal for this kind of analysis.

Key Words: Variable pressure scanning electron microscope (VPSEM), environmental scanning electron microscope, skirting, X-ray microanalysis.

*Address for correspondence:
C. Mathieu
Université d'Artois, Faculté Jean Perrin
SP 18, 62307 Lens Cedex, France
Telephone number: +33-3-21791710
FAX number: +33-3-21791717
E-mail: mathieu@univ-artois.fr

Introduction

Scanning electron microscopy (SEM) is traditionally performed in a vacuum, while the vast majority of microscopes operate at a pressure below 10^{-2} Pa. A number of manufacturers now offer instruments to perform SEM at relatively high pressure. This includes a variety of techniques reported in the scientific and commercial literature, e.g., environmental scanning electron microscopy (ESEM), Wet-SEM, controlled-atmosphere scanning electron microscopy (CAT-SEM), Low Vacuum SEM and Variable Pressure Scanning Electron Microscopy (VPSEM).

Farley and Shah (1990a,b) introduced the term high pressure scanning electron microscopy (HPSEM) to distinguish these techniques from conventional high vacuum techniques such as regular SEM and low temperature SEM. The conventional Everhart-Thornley detector cannot be used in the HPSEM, and the following modes of detection have been employed:

Specimen current mode and biased current mode

Farley and Shah (1990a,b) reported that images of a quality comparable to those obtained by the Everhart-Thornley detector could be obtained by a new detection mode. This new mode of detection was called the "bias specimen current detection mode". A biasing electrode is used above the specimen to influence the trajectories of the charge carriers and hence the image contrast. The specimen is connected, via the specimen stub, to the virtual earth terminal of a charge-sensitive amplifier to collect the current generated in and around the specimen. It has been successfully shown that the current can be collected from the specimen for the purpose of image generation, for both conducting and non-conducting specimens.

Emissive mode

The emissive mode of detection has been employed using a "gaseous detector device" [GDD, see Danilatos (1990a) for a review]. The GDD is a collecting electrode that, under working conditions is placed in the vicinity of the specimen and it is positively biased. The electrode collects emitted electrons, along with the electrons generated by the emitted and the primary electrons due to ionization processes.

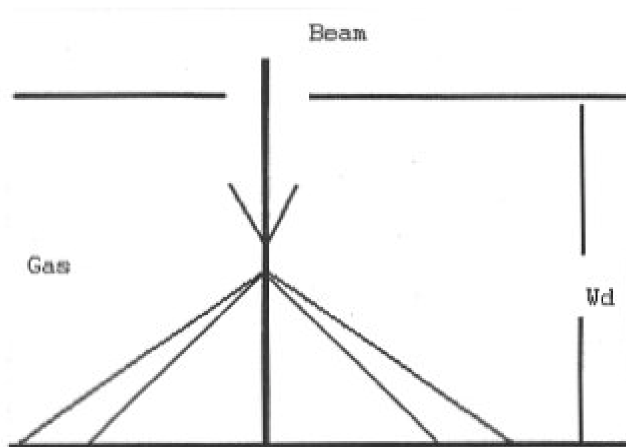


Figure 1. An electron in the beam passing the pressure limiting aperture undergoes a collision and is scattered through an angle θ.

Backscattered mode

Backscattered electron imaging with high gas pressure in the specimen chamber has proven to be a useful technique in the scanning electron microscope. For example, the specimen may be surrounded by air or nitrogen gas to inspect non-conductive surfaces (Moncrieff *et al*., 1978, 1979; Danilatos and Robinson, 1979) or by water vapor in biological applications (Robinson, 1974). This technique has been used in the Variable Pressure Scanning Electron Microscope (VPSEM) (Mathieu, 1996).

In the next section, the different kinds of interactions which take place inside the specimen chamber will be described.

Outline of General Interactions in the VPSEM

When the electron beam strikes a specimen, there is a host of reactions or interactions between the primary electron beam and the specimen, and their study has constituted a fundamental topic of electron microscopy. Thus, a primary electron may undergo elastic or inelastic collisions in the specimen resulting in the generation of secondary (SE) or backscattered electrons (BSE), X-rays etc., and changes in the specimen by molecular scission or cross linking. All of these different interactions are characterized by the fact that they occur between two entities: the beam and the specimen.

By allowing gas around the specimen, the number and type of reactions are multiplied and it is helpful if these reactions are classified and studied in a logical manner according to some natural distinction. Four main entities can be distinguished which interact with each other: beam, gas, specimen and signals (Danilatos, 1990b). Therefore, the large number of reactions can be subdivided into six general types of interactions as will be discussed below. These general types are not independent from each other and they may influence each other.

Beam-specimen interactions

Interactions of the electron beam with the specimen can result in:

(1) Beam scattering which determines the interaction volume,

(2) Generation of signals,

(3) Modification of the nature of the specimen (beam irradiation effects).

Beam-specimen interactions have constituted the primary objective of study in electron microscopy.

Beam-gas interactions

The electron beam and the gas interact with each other and the result of this interaction is

(1) Scattering of the beam,

(2) Generation of signals such as SE, BSE, X-rays and cathodoluminescence,

(3) Modification of the gas due to the creation of positive and negative ions, dissociation products and excited molecules.

The scattering of the beam will constitute the subject of a detailed analysis. This process determines the limits of contrast and resolution. The generation of signals in the gas by the primary beam should be examined in conjunction with the signals generated by the beam-specimen interactions. The signals generated by a primary beam in the gas add a constant level of noise to the corresponding useful signals from the specimen.

Specimen-signal interactions

These types of general interactions results primarily in signal modifications and to a minor extent in specimen modification. Some signals are modified by the specimen, e.g., SE are modified by a charged surface, or BSE by topographic undulations.

Signal-gas interactions

The result of signal-gas interactions is a mutual modification both of the signal and the gas. This type of interaction is of extreme significance in the VPSEM and it is an important area of investigation. Indeed, the signal-gas interactions consist of various forms of particles (including photon) collisions and have been studied quite extensively in the fields of particle physics and radiation chemistry, respectively. The effect of the gas on a particular signal and vice versa can be looked on as a first step in the chain of signal detection. This concept to use the gas as a detection medium as well as an environmental conditioning medium was introduced by Danilatos (1990a) in the ESEM.

The gas modifies the different signals to varying degrees and it is possible that the conventional detectors

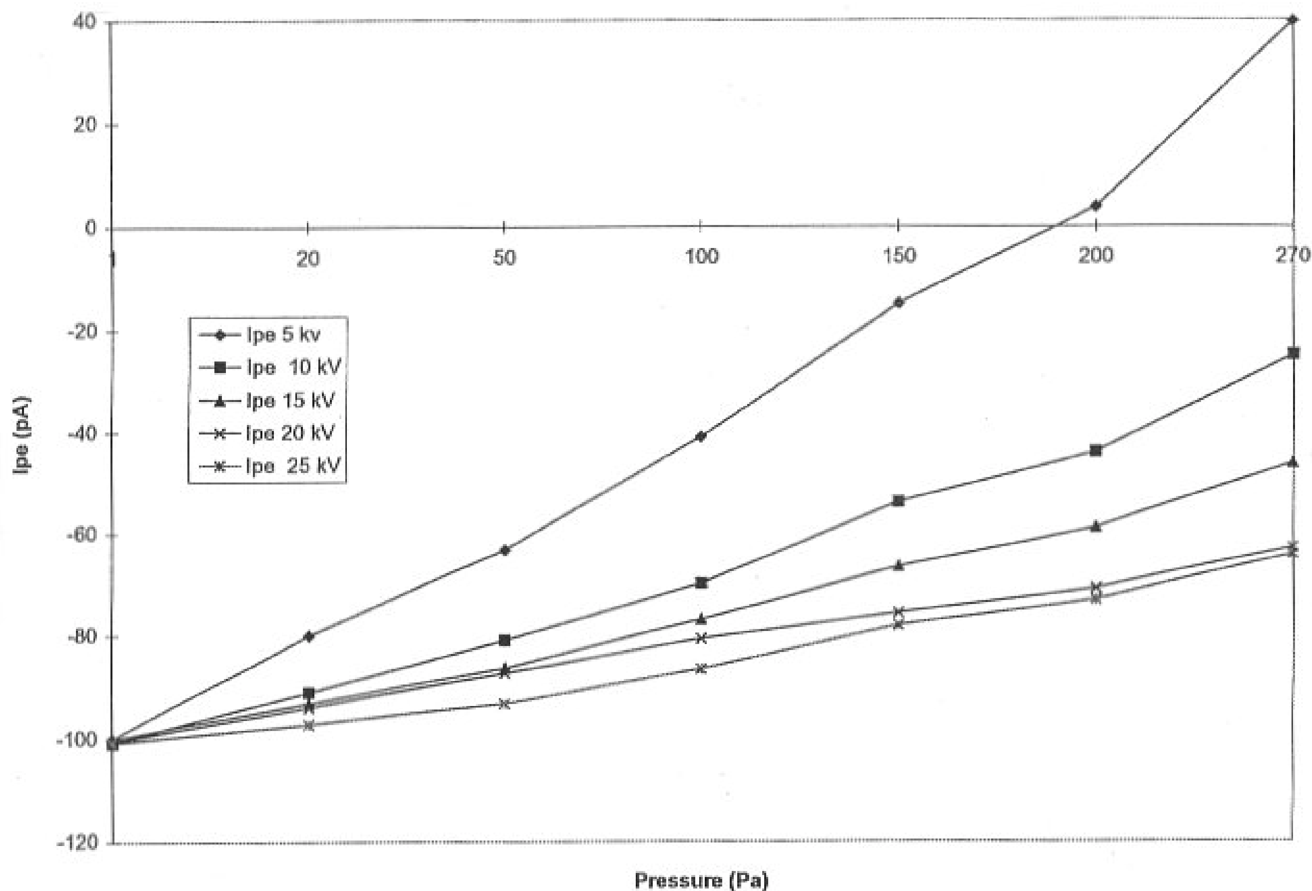


Figure 2. Variation in primary beam electron versus the pressure and versus the accelerating voltage. (Working distance = 11 mm).

(Everhart-Thornley detectors) cannot operate in the traditional form or have to be modified or new ones must be designed. The gas modification by the signals is similar to that caused by the primary beam except that it occurs over a much larger area.

Gas-specimen interactions

The gas-specimen interactions are as expected from the general physico-chemical reactions in studies outside electron microscopy. For example, the oxidation of methanol on silver particles in the ESEM has been described recently (Millar *et al*., 1997). However, the product from the beam-gas and the signal-gas interactions may modify these reactions or even initiate new reactions.

Beam-signal interactions

The last combination of the four entities suggested in the opening of this section is that of beam-signal interaction. As no practical significance can be seen at present, the direct beam-signal interaction will not be considered in this study. However, the beam can affect the signals indirectly through its interaction with the gas (background noise).

In the experimental part of this paper, the beam-gas interactions and the signal-gas interactions and the consequences for energy dispersive X-ray (EDX) micro-analysis in the VPSEM will be described.

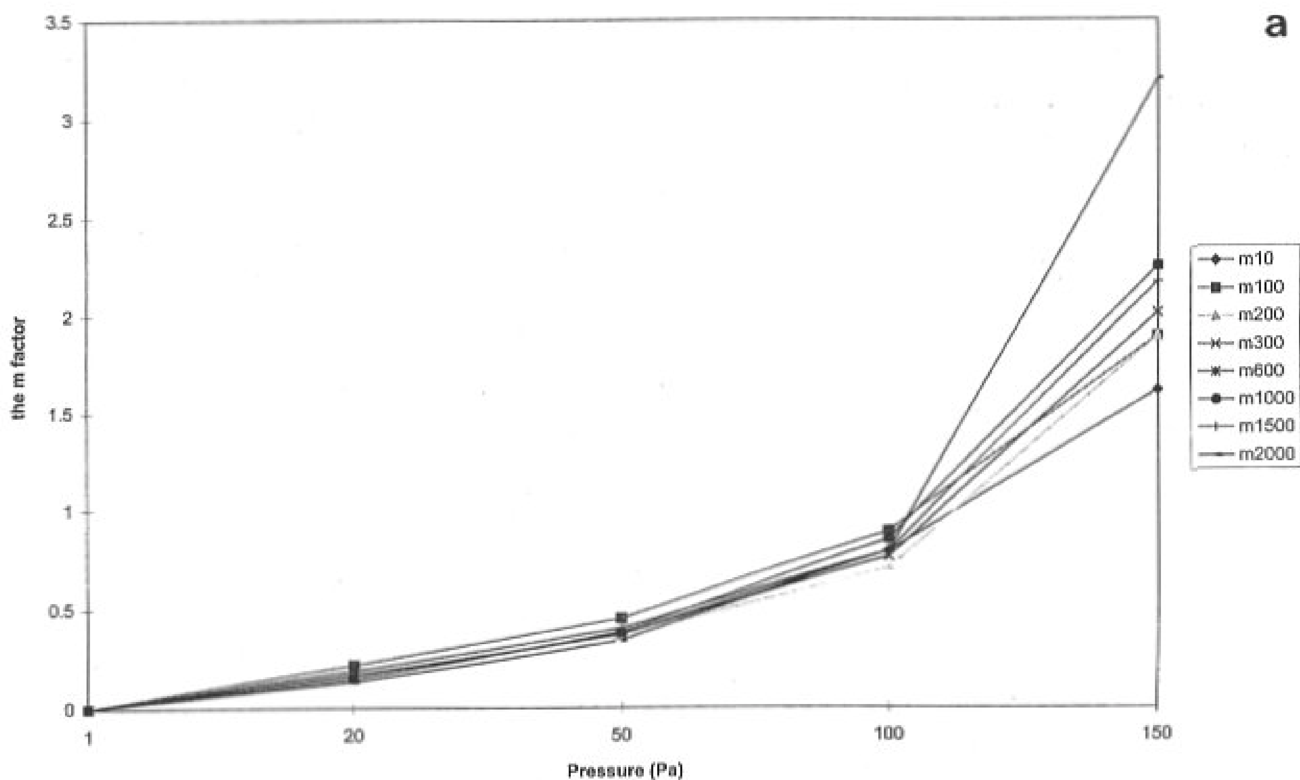
Materials and Methods

Specimen preparation

Inorganic specimens were prepared by mounting a copper target from Prolabo and an albite sample from Agar (Stansted, UK), respectively, onto an aluminum stub for measurements of the loss of spatial resolution due to beam skirting and for other EDX microanalysis experiments.

Microscopy and analysis

Studies were carried out using a variable pressure scanning electron microscope (Hitachi S2460N; Hitachi, Tokyo, Japan) with a KEVEX (Valencia, CA) atmospheric thin window. Specimen chamber pressure ranged from high



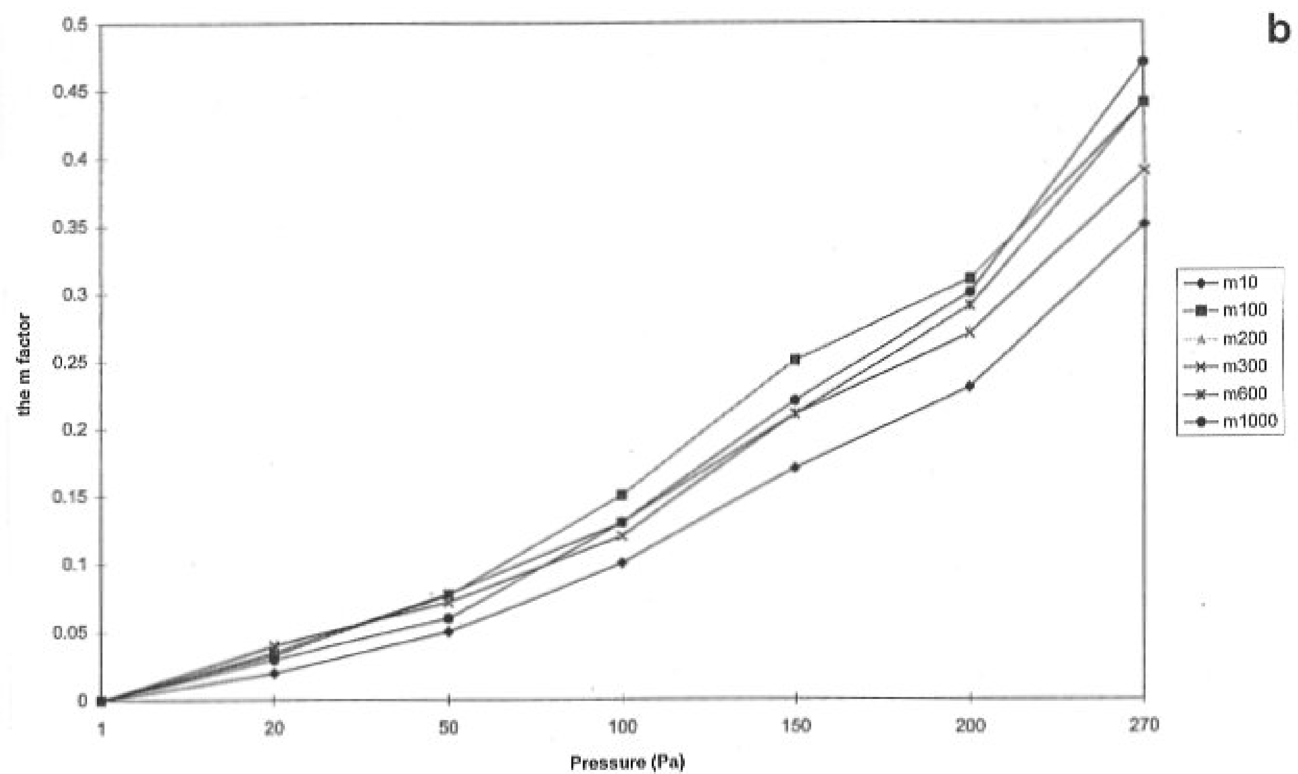


Figure 3. (**a**) Variation of the *m* value with the probe current intensity versus the pressure at 5 kV, (**b**) Variation of the *m* value with the probe current intensity versus the pressure at 25 kV.

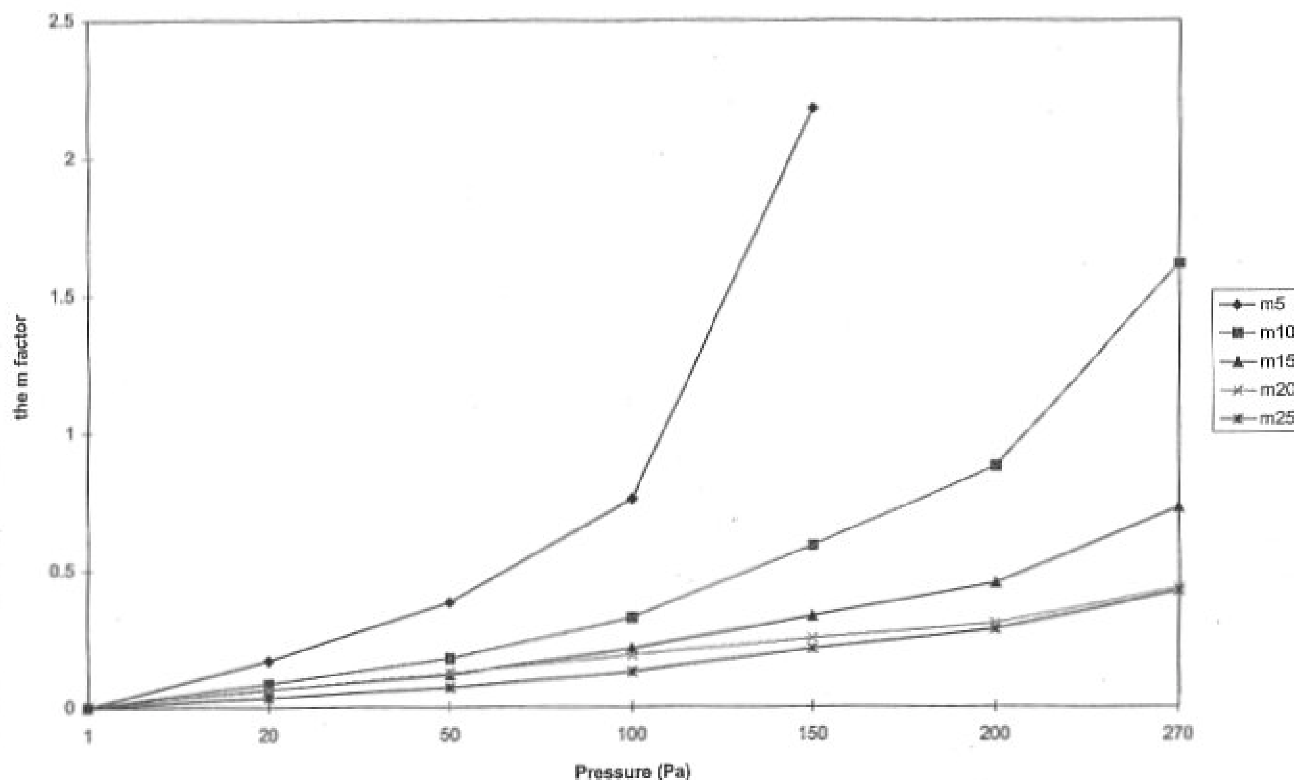


Figure 4. Variation of the *m* value at different accelerating voltages versus the pressure.

vacuum (10^{-3} Pa) to 270 Pa with an air atmosphere. The microscope was also equipped with a probe current meter (KE Development, Cambridge, UK).

With the microscope adjusted to the analytical mode, visualization of the specimen was carried out with a working distance of 25 mm. EDX microanalysis was carried with a 5, 10 and 15 kV electron probe for a live time of 100 seconds. Specimens were analyzed in the spot mode and the count rate was typically $1–2\times10^3$ cps.

Monte Carlo simulation

The Monte Carlo simulations were obtained with the Natural SEM software developed by Kimio Kanda (Hitachi). With this software, the influence of the pressure, the working distance, the accelerating voltage and the nature of the gas were investigated.

Results and Discussion

The beam-gas interaction

The electron distribution of a beam focused at a distance L below the final aperture in vacuum is modified when gas is introduced into the specimen chamber. It is of fundamental importance to know the details of the new electron distribution resulting from the collisions of electrons with gas molecules or atoms. A collision occurs when the electron passes within a characteristic area around the particle known as the total cross section σ_T. With each collision, the electron may loose some energy ΔE and become scattered at an angle θ away from the initial direction. This effect is called skirting (Fig. 1).

The influence of the accelerating voltage, pressure and the nature of the gas on the skirting effect was investigated. The probe current was adjusted at a fixed value at 10^{-3} Pa. Figure 2 shows probe current variations versus the pressure at 5, 10, 15, 20 and 25 kV. The scattered part of the electron beam increases with the increase of the pressure and the decrease of the accelerating voltage, respectively. These variations are characteristic of the influence of a gaseous environment. In order to analyze the beam gas effect, a factor *m* which is the average number of collisions per electron is introduced. If the intensity of electron beam in vacuum is I_0, the fraction of beam I transferred intact is given by:

$$I = I_0 e^{-m} \quad (1)$$

thus it is easy to determine experimentally the value of *m* for each pressure. Figures 3a and 3b show the variations of

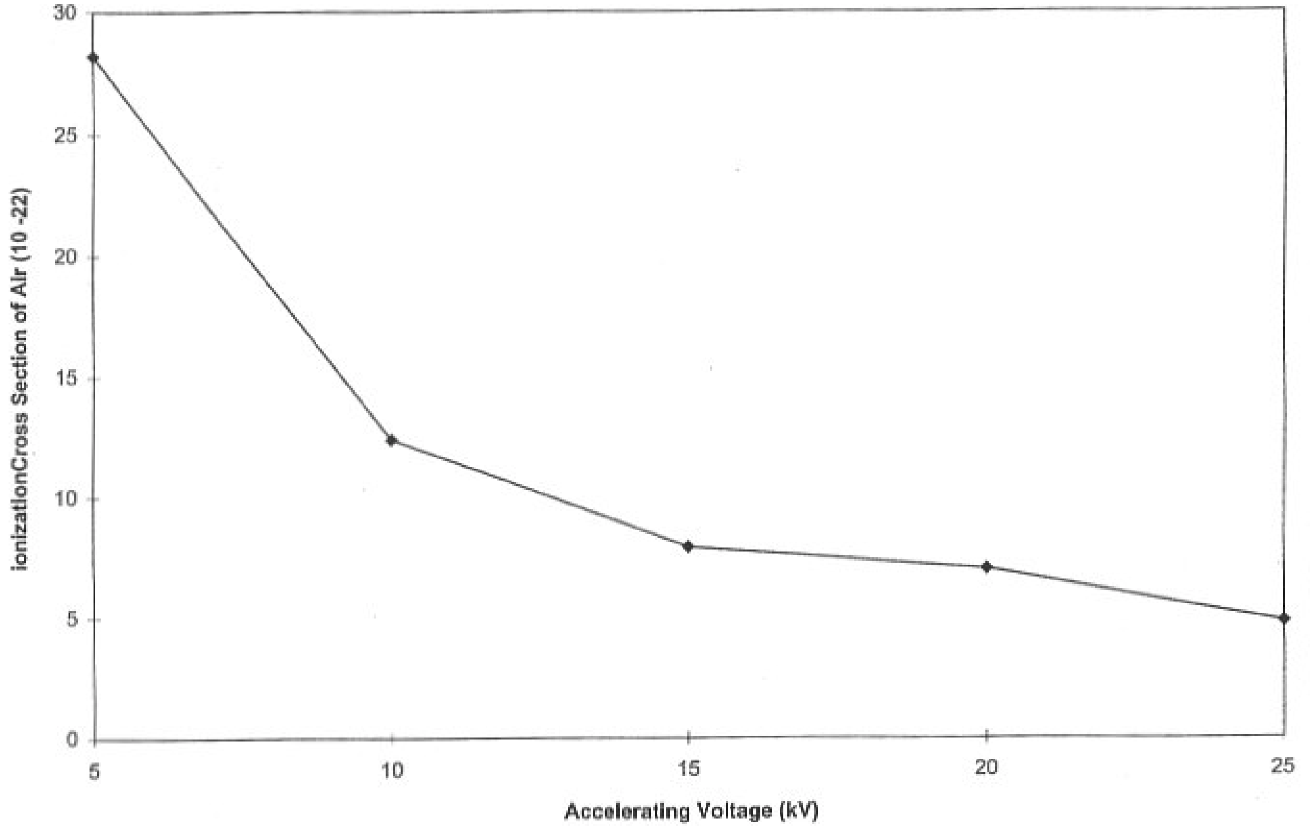


Figure 5. Variation of the ionization cross section versus the accelerating voltage.

the value of m as a function of the pressure for different intensities of the primary beam which varied from 10 to 2000 pA at 5 and 25 kV. A linear dependence is obtained between m and the pressure at 25 kV. Moreover, the probe size does not influence the beam gas effects except in the case of 5 kV between 100 to 150 Pa where the m value is very important, which indicates that the relationship which defines the m value is correct until 150 Pa, after the current becomes positive. The average number of collisions is also defined by Danilatos (1990c):

$$m = \sigma_T \, p \, L / k \, T \qquad (2)$$

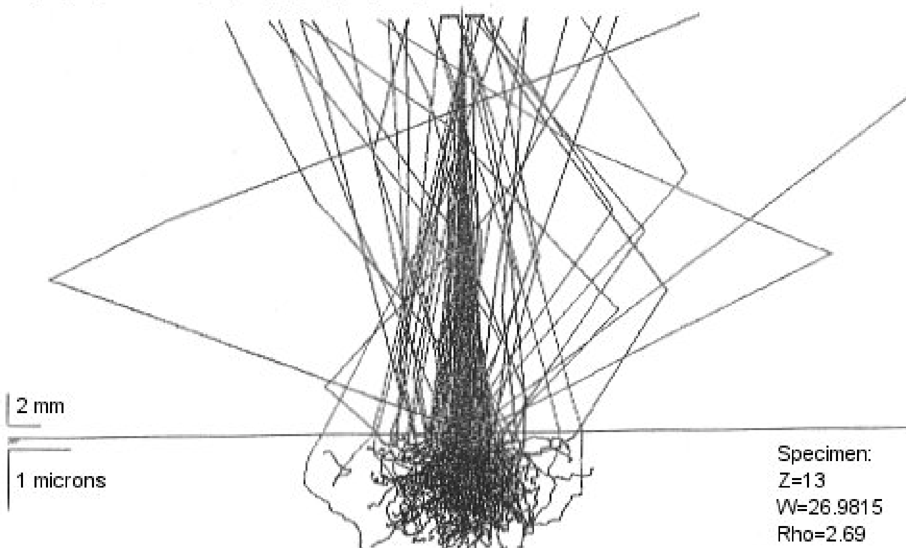
where σ_T is the scattering cross section for the particular gas used (m^2), p the pressure (Pa), L the working distance (m), T the temperature (K) and k the Boltzman constant ($1.38\ 10^{-23}\ JK^{-1}$). This relationship indicates that the number of collisions increases with the pressure, thereby corroborating these results.

As the probe size does not influence the results, Figure 4 shows the variations of the calculated value of m for different intensities of the accelerating voltage. For each accelerating voltage, the slope can be used to determine the total ionization cross section. In the range between 1 to 100 Pa, Figure 5 represents the variation of the total cross section ionization versus the accelerating voltage. The shape of the curve agrees with the results obtained by Farley and Shah (1990a).

It is difficult to compare the experimental value with the literature because for the range of energies used; the total cross section ionization for the most commonly used gases such as H_2O or O_2 is not readily available. However, this method can allow the easy evaluation of the total scattered ionization cross section for different gases.

Correlation with Monte Carlo simulations

The interaction between gas and electrons can be investigated with a Monte Carlo simulation. Figure 6a represents the beam-gas interaction in an air atmosphere at 5, 15, or 25 kV. This simulation shows the fact that the ionization cross section is very important at low voltage. Therefore, the skirting is very important at low voltage. In addition, Equation (2) also indicates the influence of the working distance which is illustrated in Figure 6b and of the pressure (Fig. 6c). The experimental results are in good agreement with the different simulations. Moreover, it is interesting to consider the effect of the nature of the gas. Three examples (helium gas, air, and krypton) are given in Figure 6d. The simulations show an important effect of the



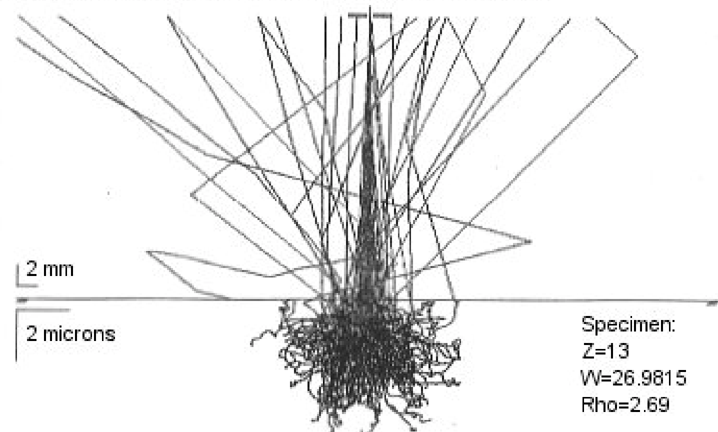


Figure 6. Simulation of the skirting effect. (**a**) Influence of the accelerating voltage (E_0 = 5, 15, 25 kV) (Working distance = 25 mm, P = 1 torr air).

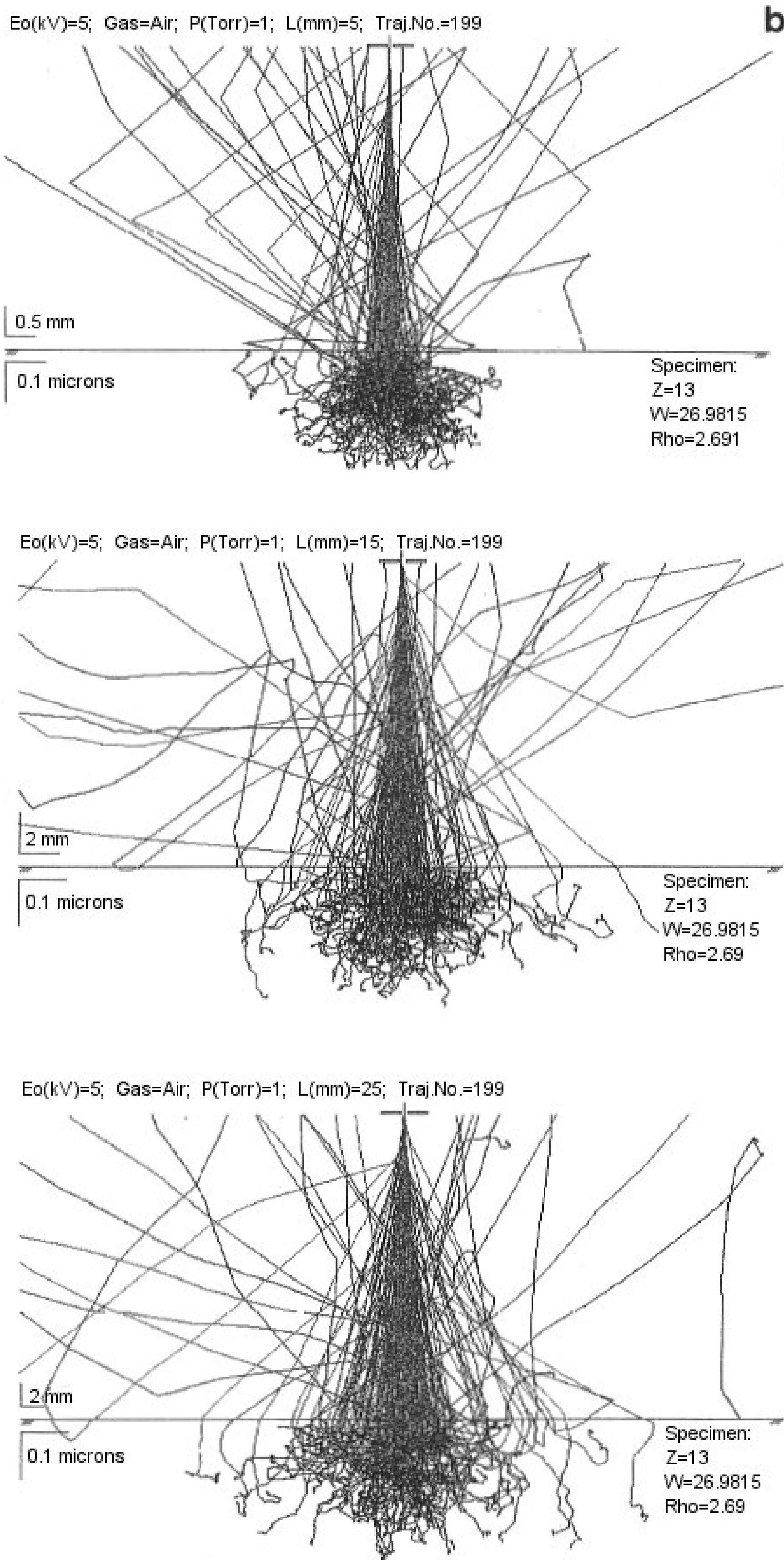


Figure 6. Simulation of the skirting effect. (**b**) Influence of the working distance (5, 15, 25 mm) (E_0 = 5 kV, P = 1 torr air).

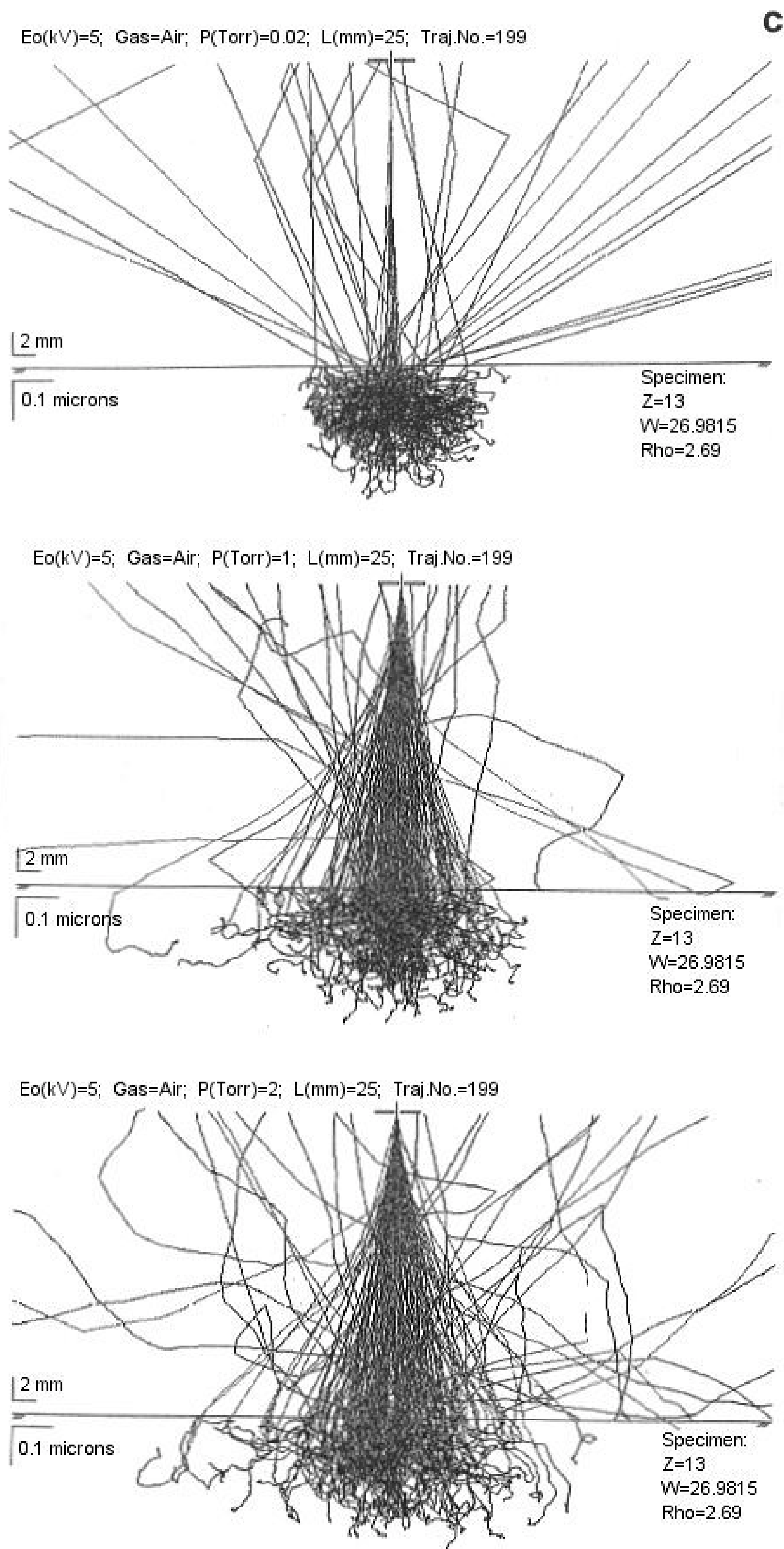


Figure 6. Simulation of the skirting effect. (**c**) Influence of the pressure (P = 0.02, 1, 2 torr) (E_0 = 5 kV, Working distance = 25 mm, air).

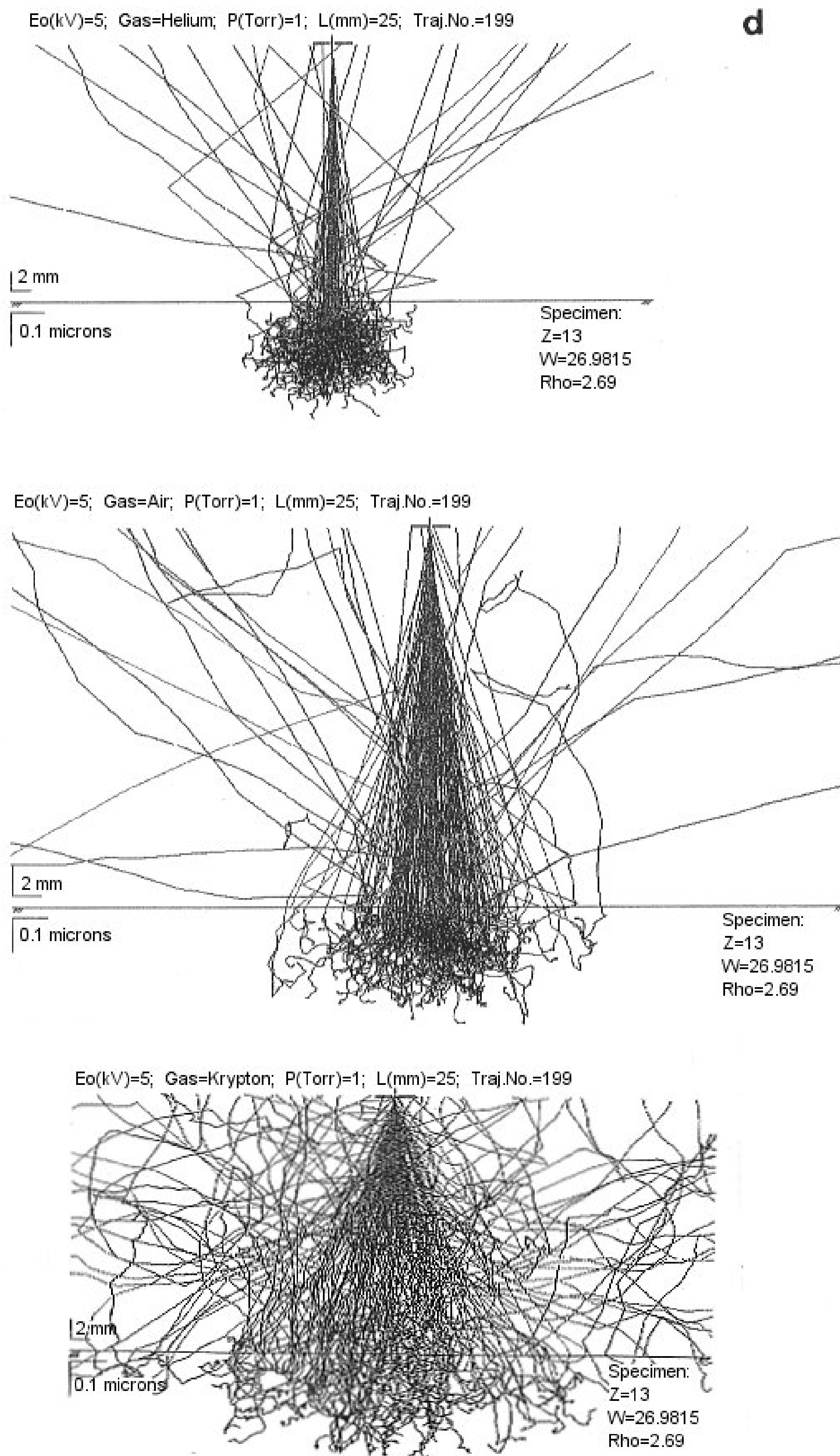


Figure 6. Simulation of the skirting effect. (**d**) Influence of the nature of the gas (He, Air, Kr) (Working distance = 25 mm, E_0 = 5 kV, P = 1 torr).

nature of the gas on beam skirting and the choice of a gas with a low atomic number is recommended in order to reduce the skirting effect in the VPSEM.

Signal-gas interactions

The sample (gold on carbon) was imaged at a magnification of 80000 as a function of the pressure at 1, 20, 50, 100, 200 or 270 Pa at a working distance of 11 mm with an accelerating voltage of 25 kV. The primary beam current was measured in order to estimate the skirting. In these conditions, the value of m varies from 0 at 1 Pa to 0.2 at 270 Pa.

The different images (Fig. 7) reveal that the signal is sufficient to obtain a correct image even at 270 Pa. However, the signal-to-noise ratio decreases and consequently the image quality decreases at high pressure. The scattered part of the electron beam at 270 Pa represents 20 % of the initial value. Therefore, the emission of the backscattered electrons at the impact point is reduced and the interaction between the gas and the primary electrons and also the interaction of the emitted backscattered electron with the gas add a constant level of noise to the corresponding useful signals from the specimen. The signal-gas interactions disturb the image by increasing the level of noise. It will be interesting to carry out the same kind of experiment with a gas with a low atomic number in order to reduce the interaction between the gas and the beam.

Consequences for microanalysis

The presence of a gaseous environment in the specimen chamber presents serious limitations for the use of X-ray microanalysis (XRMA) in the VPSEM due to three major effects: (1) *loss of spatial resolution* due to beam skirting (beam-gas interactions), (2) *contribution of X-rays from the gas atmosphere* elicited by the primary electron beam and by backscattered electrons (signal-gas interaction), and (3) *a reduced X-ray count* (beam-gas interactions).

Loss of spatial resolution due to beam skirting

Sigee and Gilpin (1994) proposed a simple experiment in the ESEM to demonstrate the beam skirting which had been reproduced in the VPSEM. An electron beam was directed onto an aluminum stub with a copper target placed at varying distances from the primary probe area. The skirting effect of the beam in an air atmosphere was shown by the ability to pick up a Cu signal of varying intensity from the central probe area (Fig. 8).

Most data were obtained at distances of 100-1000 mm from the Cu target. At high vacuum, no Cu peak was detected at these distances. At a distance of 1 mm, the copper content varied from 2 to 9 % when the pressure ranged from 1 to 100 Pa (Figs. 9a and 9b) at 15 kV.

The Cu peaks were always detected over the 100-1000 mm range between 1 to 100 Pa, indicating a clear skirting effect in the presence of an atmosphere. Beam skirting clearly limits the spatial resolution of X-ray microanalysis. According to the results obtained in the previous section, the spatial resolution can be improved with the use of high accelerating voltage and low working distance.

X-ray contribution from the chamber atmosphere

Passage of the electron beam through the specimen chamber atmosphere leads to the generation of characteristic and continuum X-rays from the gas molecules. In order to illustrate this effect, a sample which contained Na, Al, Si and O was analyzed at three different accelerating voltages (5, 10 and 15 kV). With the same excitation conditions, the effect of introduction and increased levels of air atmosphere was investigated by collection of X-ray from the sample. X-ray emission spectra at high vacuum indicated only peaks of O, Na, Al and Si.

Figure 10 shows the variation of the ratio of the aluminum content at different pressures to the aluminum content at 1 Pa. For the same three accelerating voltages, an increase of the ratio was observed with increasing pressure. The effect of the skirting is more important at low accelerating voltage which corroborates the previous results. Figure 11 shows the variations of the ratio of the oxygen content at different pressures to the oxygen content at 1 Pa which is used as a reference, at three different accelerating voltages. The curve obtained at 5 kV shows an important increase of this ratio. This behavior is not observed at 15 kV. The air atmosphere in the specimen chamber gives rise to the generation of an oxygen signal, which increased with the atmospheric pressure. The presence of an atmospheric X-ray contribution clearly presents problems for the determination of the oxygen content in the analyzed specimen. The X-ray contribution from the atmosphere clearly limits quantitative X-ray microanalysis. Various operational parameters can be optimized to reduce this effect including the atmospheric pressure and the nature of the atmospheric gas.

Atmospheric pressure. The chamber pressure should be kept as low as possible.

Nature of atmospheric gas. In order to reduce the X-ray contribution from the atmosphere, a practical solution is to choose a gas such as H_2 or He. The atmospheric contribution from these gases will not be detected by XRMA.

Reduced X-ray count

Introduction of an atmosphere into the specimen chamber can lead to a marked fall in the total counts in the X-ray spectrum. In the VPSEM, with the same excitation conditions, air atmosphere causes a significant reduction of the total X-ray counts (Fig. 12). This effect is more important at low accelerating voltage. The decrease of the total counts can be explained by a reduced electron beam penetration to the specimen due to electron scattering and also probably

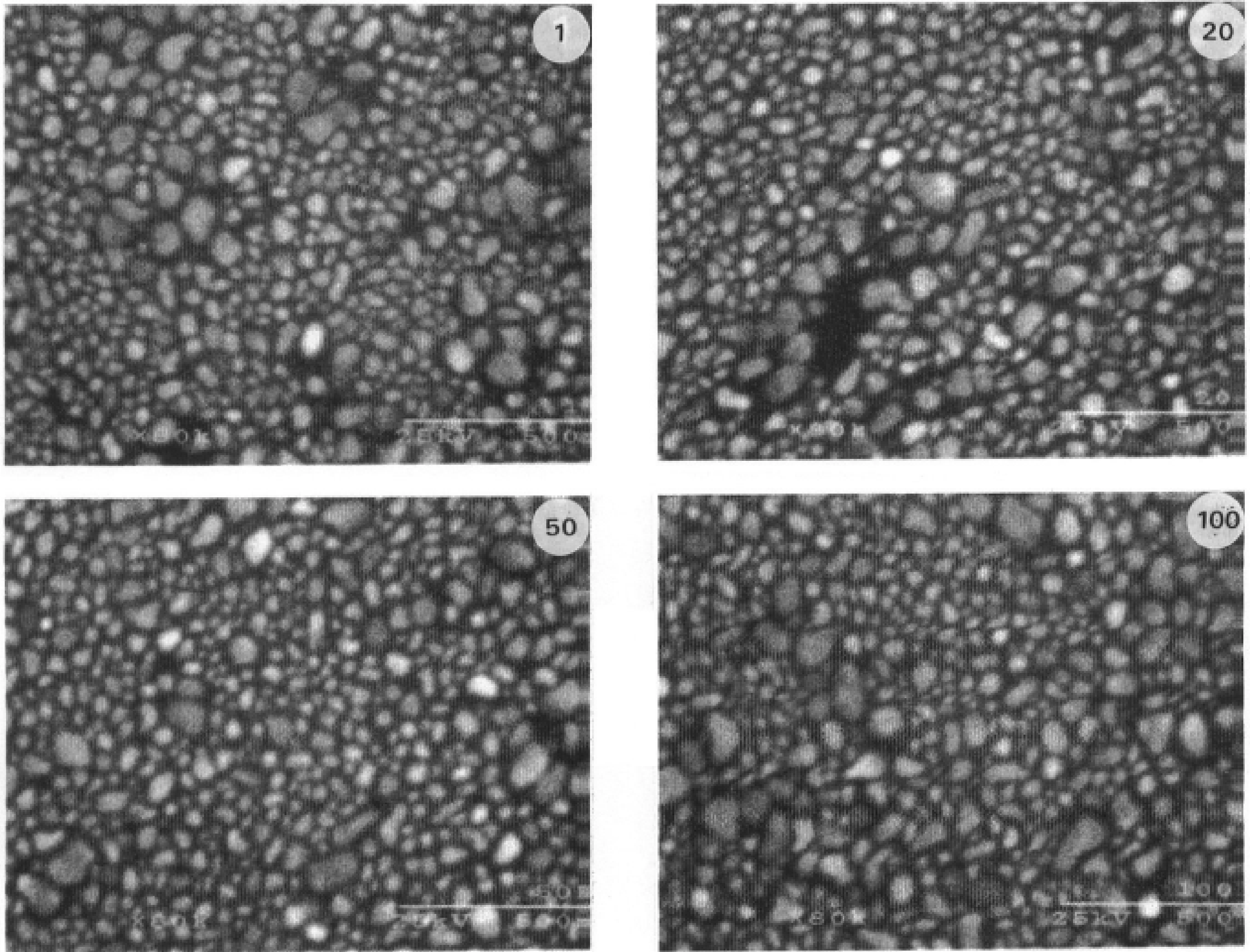


Figure 7. Variation of the image quality in function of the pressure (1, 20, 50, 100, 150, 200, 270 Pa) at 25 kV (*continued on facing page*).

by the absorption of X-ray photons in the gas at high pressure. In order to reduce this effect, the choice of a gas with a low atomic mass such as hydrogen or helium gas can be an interesting solution.

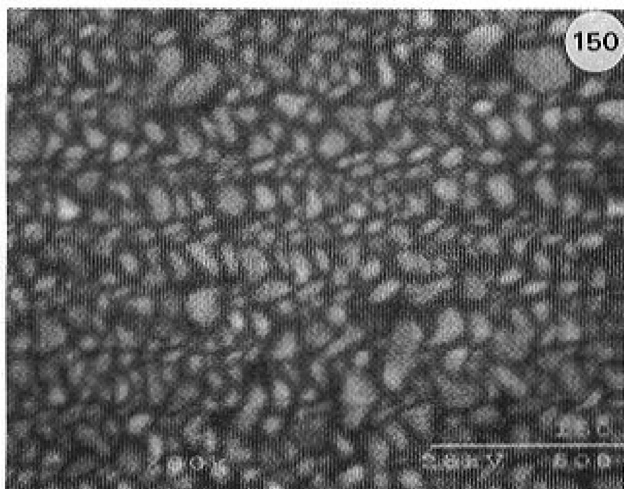
The correction methods for X-ray microanalysis

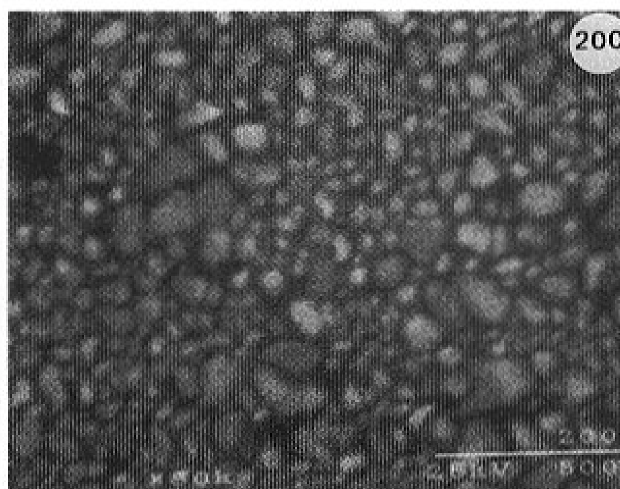
In order to take into account these effects in quantitative X-ray microanalysis (Sigee, 1998), different correction methods exist. Two basically different correction methods have been proposed by Bilde Sorensen and Appell (1996) and are referred to as the beam stop and pressure variation procedures, respectively.

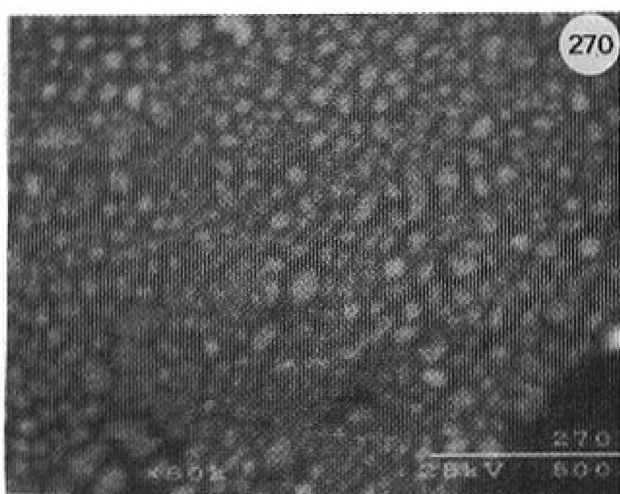
Beam stop method. One version of this procedure involves the use of a needle (composed of a known element not present at detectable levels in the sample) that can be inserted over the specimen to act as a beam stop for the central (unscattered) electrons but not for the peripheral beam skirt. The spectrum obtained with the beam stop in position contains X-rays derived from the known element (beam stop) plus the area covered by the beam skirt. The pure spectrum from the central probe area can be obtained by removing the characteristic peaks of the known element from the first spectrum and subtracting the remaining spectrum from the second.

Pressure variation method. This approach is based on the fact that the intensity of the skirt varies with chamber pressure, and thus correction for electron scattering can be made by obtaining x-ray spectra at different chamber pressures. The expression can be used to relate the measured count rate (C_T) from a particular element in the sample to count rates at zero scattering (C_u) and complete scattering (C_s).

$$C_u = C_u \exp(-m) + C_s [1-\exp(-m)] \quad (3)$$







where $m = p\,s\,L/kT$ as previously defined.

The factor C_s is unknown, but will be a constant provided that p*m* is sufficiently low for multiple scattering of electrons to be ignored. Under these conditions, C_u can be derived from two measurements of C_T at different pressures, where *m* is known.

Another correction method has been proposed by Doehne (1997). This method is as follows: an X-ray spectrum (A) is acquired under condition of high chamber pressure (2P). Another spectrum (B) is made under identical conditions but at a lower chamber pressure (P). The difference between the two spectra provides information on how the decrease of the contribution of the X-rays generated by the skirt electrons affects the overall spectrum. If C is the spectrum at low pressure ($<10^{-3}$ Pa, no skirt effect), then C can be approximated by the following:

$$C = B - [(A-B)*D] \qquad (4)$$

This methods assumes that changes in the lateral extent of the X-ray skirt with pressure are less important than changes in the skirt intensity. D is an empirical factor derived from observation when the background shape of spectrum B is significantly altered by the subtraction of the spectral differences between A and B. The background shape acts as a built-in check against overcorrection. It is important to notice that the correction methods described above only account for the skirting effect.

Conclusion

The beam gas interaction is very important in the VPSEM. The skirting effect can be reduced if the instrument operates at low working distance and high accelerating voltage. The choice of a gas with a very low atomic mass can provide an interesting solution in order to reduce the skirting. The spatial resolution can be improved by optimizing these parameters. However, the use of a high accelerating voltage for the analysis of light element is not optimal. Therefore, the choice of atomic gases with low molecular weight will be certainly the best solution. The future development for quantitative X-ray microanalysis may involve the introduction of correction factors that take into account not only the skirting effect, but also the atmospheric contribution.

Acknowledgements

The author would like to thank Thierry Grenut from Elexience (Verrieres le Buisson, France) for technical assistance and Kimio Kanda from Hitachi for the use of the software natural SEM.

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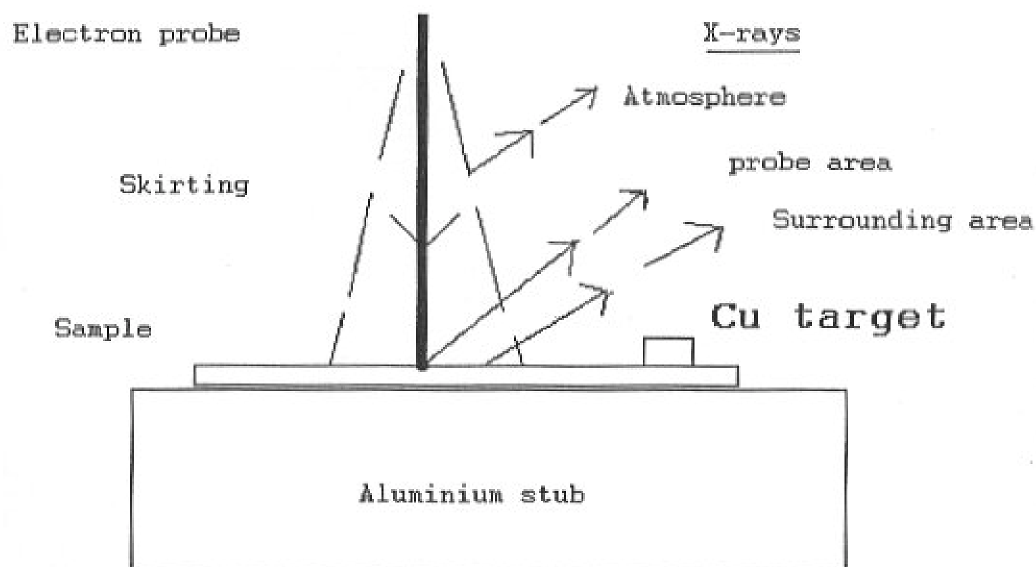

Figure 8. Demonstration of the beam skirting using a Cu target (from Sigee and Gilpin, 1994).

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Discussion with Reviewers

B. Breton: Is the sole reason for using a VPSEM to eliminate charging? The evident problems with quantitative microprobe analysis suggest that the conventional application of a thin conductive film will enhance both resolution and quantitative accuracy; so why go to all this trouble? Perhaps the authors would clarify why a VPSEM was used.

Author: The VPSEM allows observations to be carried in the presence of gas at pressures of up to about 2 Torr in the specimen chamber. This arrangement makes it possible to image many kinds of samples that would be unsuitable for a conventional SEM because they are dirty, moist or even wet. In addition, poorly conducting or insulating samples can be imaged at high beam energies (typically 10-30 keV) without the need for a conductive coating because the ionizations occurring in the gas as the result of electron interactions produce a flux of positive ions which migrate to charged regions and neutralize them. The fact that VPSEMs now account for over 50% of the market for conventional scanning electron microscopes proves the appeal of this concept. Therefore I think that it is important to show the various consequences of carrying out X-ray

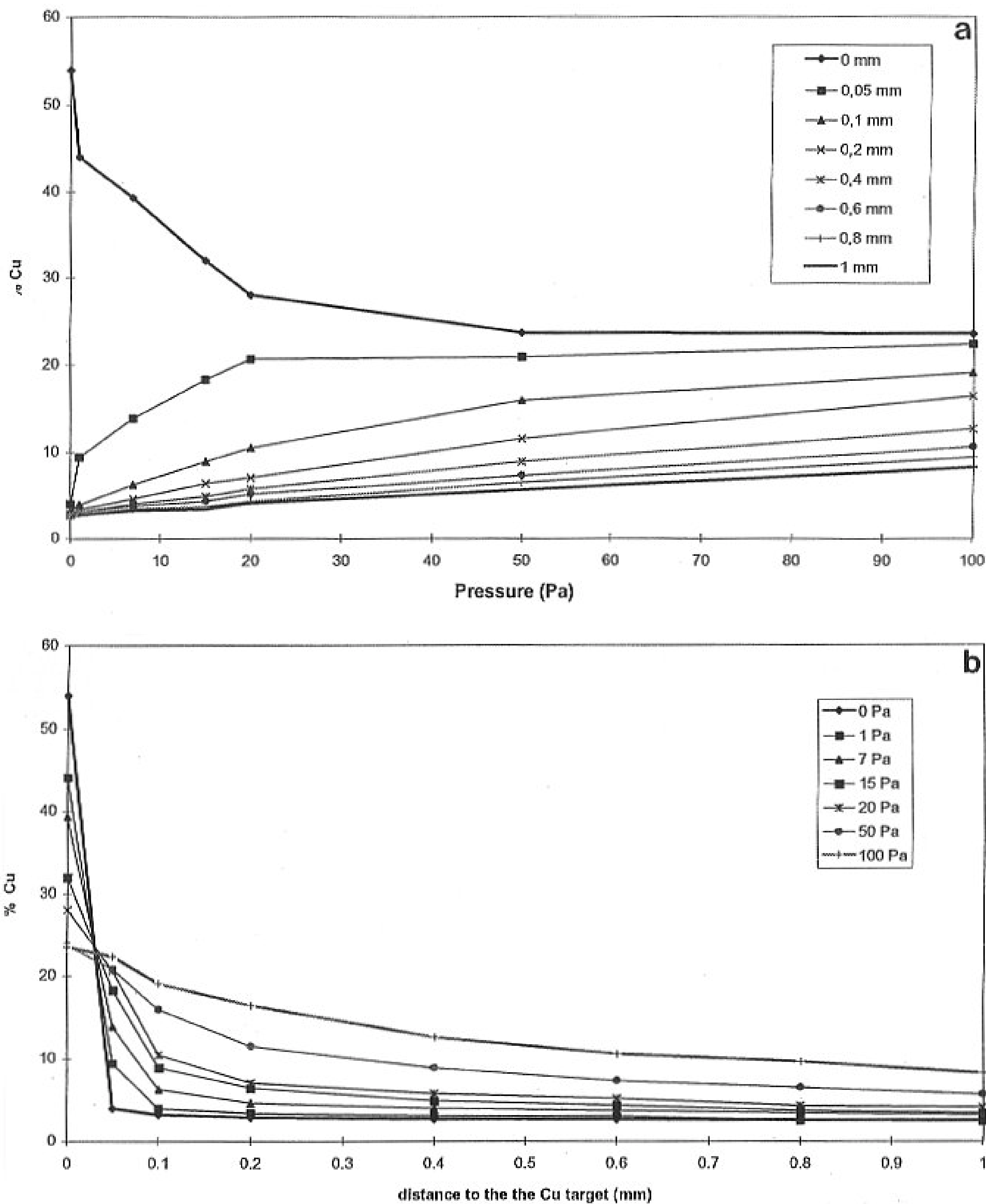
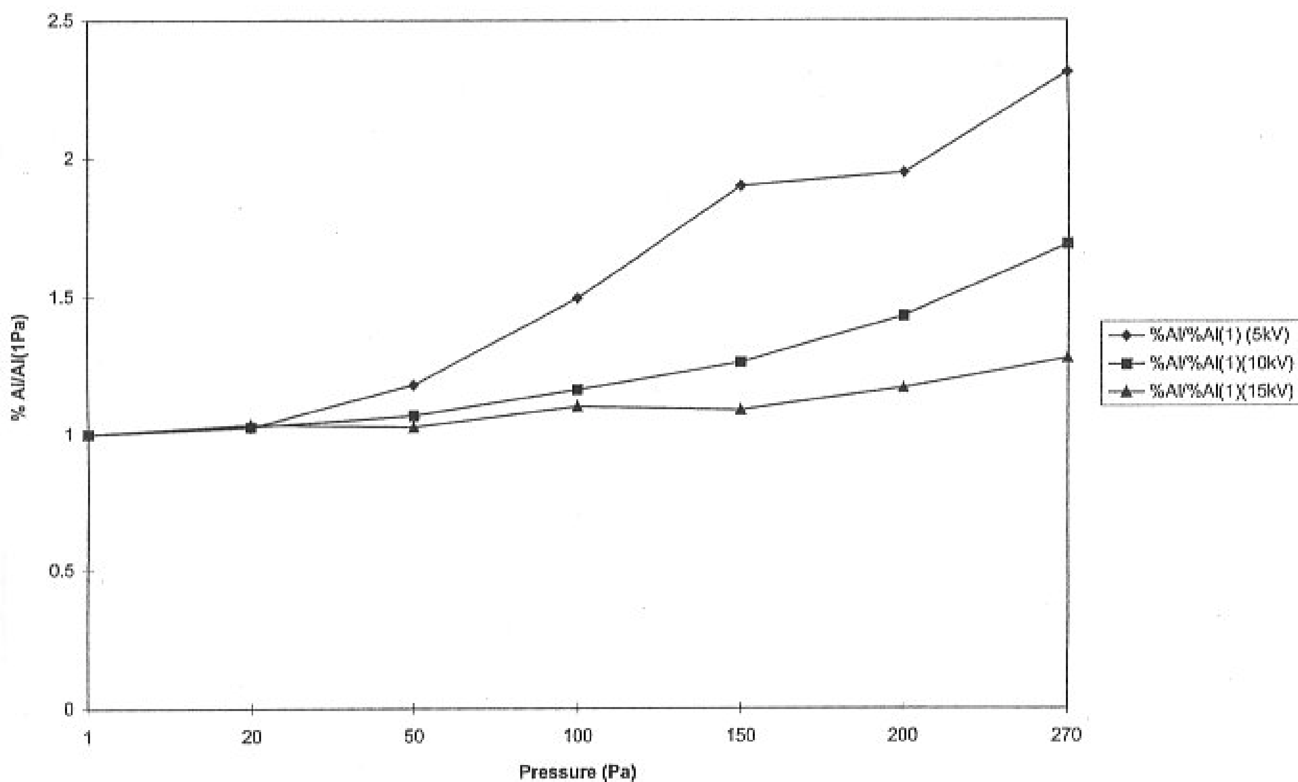


Figure 9. (**a**) Variation of the Cu content versus the pressure, (**b**) Variation of the Cu content with the distance to the Cu target.



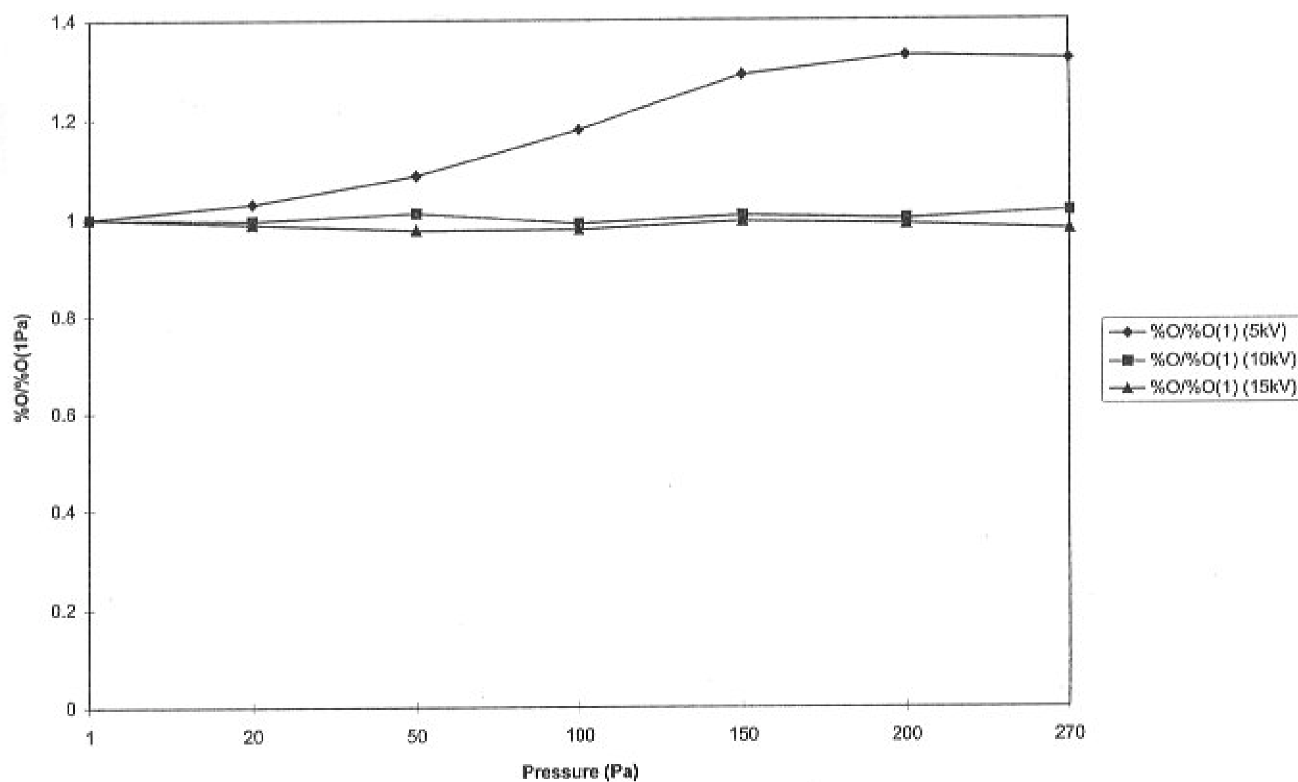


Figure 10 (*top*). Variation of the %Al / %Al (1 Pa) ratio with the accelerating voltage.
Figure 11 (*bottom*). Variation of the %O/ % O (1 Pa) ratio with the accelerating voltage.

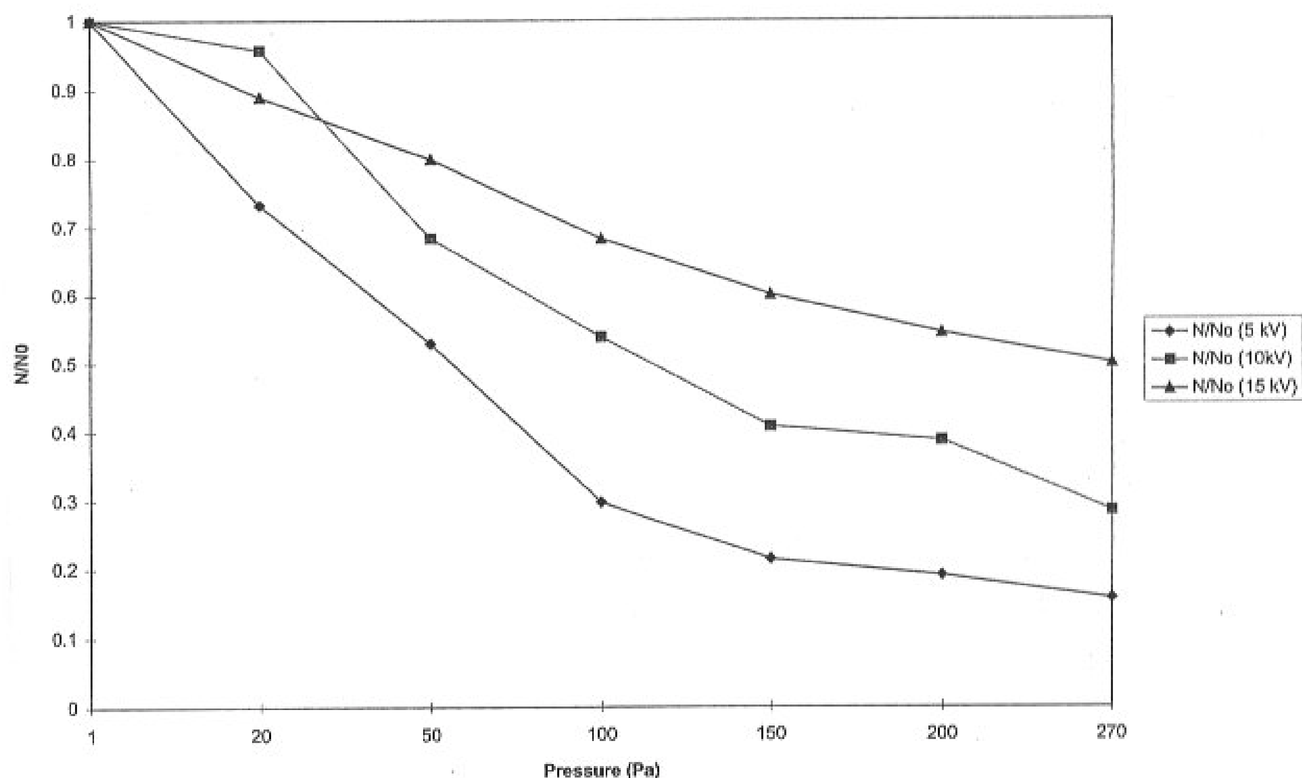


Figure 12. Variation of the ratio N/N_0 (1 Pa) with the accelerating voltage.

microanalysis in the presence of gas at pressures of up to about 2 Torr. I agree with the reviewer that it is preferable to coat the sample with a thin conductive film in order to enhance the resolution and quantitative accuracy in the high vacuum mode (10^{-3} Pa) but most operators use the VPSEM in the low vacuum mode and it is necessary to clarify the limitations of the X-ray microanalysis in the low vacuum mode.

B. Breton: Regarding X-ray contribution from atmosphere chamber: in selecting a specimen which contained the interfering elements (Al and O), is there not a danger of confusing, e.g., penetration effects within the sample at various kV with effects due to the chamber or atmosphere? If this investigation were performed using a specimen with no Al or O content, then extraneous peaks would be more positively identified.

Author: The choice of a sample containing Al, Na, O and Si deposits on an aluminum stub with the interfering aluminum element was made deliberately in order to illustrate the various effects. It is evident that the operator will choose a stub without an interfering element. Moreover, a majority of VPSEM users use air gas inside the specimen chamber; in that case, the atmospheric contribution will be apparent and the analysis of oxygen, at low kV, will be compromised. Therefore, the operator should change the gas inside the specimen as proposed by Stowe and Robinson (1998) and use helium inside the specimen chamber (elimination of the atmospheric contribution). This solution has been tested with the current measurement method (Adamiak and Mathieu, submitted for publication) in order to evaluate the decrease of the skirting effect and the results are very promising.

B. Breton: Regarding the reduced X-ray count at increased pressure: if this is due to absorption of the emitted X-ray by the gas, then I think it should be independent of kV, since only the number of X-rays emitted changes with kV; their characteristic energy remains constant. If this is so, it should be possible to separate the two mechanisms proposed by the author.

Author: The experimental results show a significant decrease of the ratio N/N_0(1 Pa) (Fig. 12) with increasing pressure and decreasing accelerating voltage. For a given accelerating voltage, the variation of this ratio is due to the increase of the beam skirting which implies a beam loss at the impact point and so the emission of X-rays is reduced. For a given pressure, the variation of this ratio with the accelerating voltage also shows the importance of the beam skirting which increases with decreasing accelerating volt-

age. As suggested by the reviewer, the reduction of the total X-ray count is the direct consequence of the beam skirting, and the reduction of the emitted X-rays is due to the reduction of the beam penetration inside the specimen chamber. This effect is probably the most important in order to explain the experimental results.

I. Müllerova: What kind of specimen is used for the simulations in Figure 6? Is it aluminum as indicated in the figures (Z= 13)?
Author: Yes, the sample for the simulation is indeed aluminum because in a previous paper (Mathieu, 1998) I studied the variations of the specimen current in aluminum samples in the VPSEM.

I. Müllerova: Could you please summarize any possible important applications of variable pressure SEMs not covered as yet? What are the main limitations of the imaging?
Author: It is difficult to summarize any possible important application of the VPSEMs not covered as yet. Indeed, the types of applications that the VPSEM is able to tackle are virtually unlimited. Unlike the conventional SEM, which is restricted to clean, dry specimens, the VPSEM can be used to observe a wide variety of wet, oily, and non-conductive specimens. The following are examples of specimens that can be observed with the VPSEM in the low vacuum mode: (1) insulating materials such as ceramics, plastics, polymers, synthetics fibers, powders, paper products, textiles, (2) plants, food, soil, insects, tissues, petroleum products, (3) hydrated cement, geological samples, (4) forensic and archeological items where sample preparation is not possible. However, I believe that an important step for the high pressure SEM will be the development of quantitative corrections for X-ray microanalysis. The principal limitation of the imaging in the VPSEM is that it cannot be used to produce secondary electron (SE) images. This is firstly because the mean free path for SE in a gas at a pressure of 0.5 Torr is only a few mm, and so none of them would reach a detector placed a few centimeters away. The absence of a true SE detector deprives the operator of the familiar benefits of mechanisms such as topographic contrast and leads to an unwelcome reduction in the contrast of surface details. Recently, in order to overcome these limitations, Mohan *et al*. (1998) proposed a solution in the VPSEM to obtain SE imaging with the system described by Farley and Shah (1990a,b) and they obtained good SE images.

D.C. Sigee: The author mentions three modes of detection in the HPSEM. Could he comment on the relative merits of these in relation to the different atmospheric pressures used in the specimen chamber?
Author: I think that for all the detectors the signal to noise ratio decreases with the increase of the pressure and the consequence is a loss of the image quality. In this paper, the loss of image quality for the BSE detector has been shown. In order to improve the image quality, I think that a solution may be to measure the specimen current and to determine the value of the pressure which corresponds to a specimen current equal to zero. Indeed, this point would be optimum point for imaging providing charge balance at the lowest possible pressure. An interesting study about the GSED (gaseous secondary electron detector) in the ESEM has been performed by Fletcher *et al*. (1997). These authors present quantitative data on the amplification behavior in different gases. They investigated water vapor, nitrous oxide, carbon dioxide and helium. It was shown that each gas had a distinctive amplification behavior. The profiles, in all cases except helium, have the same shape. This study provides quantitative information that can be used to determine the optimum conditions with regard to the nature of the gas, the primary beam energy, the detector gap distance, and the pressure. An important fact is the particular behavior of the helium gas which can be explained by the fact that the ionization efficiency cross sections for SE and BSE are approximately an order of magnitude smaller than those for water vapor. For the detector described by Farley and Shah (1990a,b), Durkin and Shah (1991) and Mohan *et al*. (1998) obtained images which closely replicate those available in a conventional high vacuum SEM. More generally, high pressure SEM techniques are being developed to meet the following objectives: (1) preservation of the integrity of the specimen under examination; (2) minimizing artefacts and maximizing the signal to noise ratio. Therefore, the conditions of operation will be dictated by the type of the specimen under consideration. The type of gas and actual operating pressure must be chosen to prevent the loss of volatile constituents of the specimen. For example, in order to preserve a fully hydrated biological system it must be kept at saturated water vapor pressure at ambient temperature. The saturated vapor pressure is a function of the temperature. It is possible to lower the vapor pressure by keeping the sample at a low temperature whereby evaporation of water is suppressed. The introduction of a specimen cooling stage coupled with high pressure SEM techniques offers new possibilities in the area of biological sample analysis.

D.C. Sigee: In Figure 4, which plots the *m* value against atmospheric pressure, you state that the slope of the graph can be used to determine the total ionization cross section. Could you elaborate further on this?
Author: With this method it is easy to determine the total ionization cross section. This value can be used in the correction method proposed by Bilde Sorensen and Appell (1996) called the "Pressure Variation Method". The expression can be used to relate the measured count rate (C_T) from

a particular element in the sample to the count rate at zero scattering (C_u) and complete scattering(C_s) as given in equation (3) above. In this equation, C_s is unknown, but will be constant provided that *m* is suffiently low for multiple scattering of electrons to be ignored. Under these conditions, C_u can be derived from two measurements of C_T at different pressures, where *m* is known. The curve of *m* as a function of the pressure is also interesting because it limits the field of application of this correction method for each pressure and accelerating voltage. Indeed, the field of application supposes that the *m* value should be kept below 0.35 if the number of scattering events has a Poisson distribution.

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