Monte Carlo simulation of the electron beam scattering under gas mixtures environment in an HPSEM at low energy

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The effects of various gas environment used in high pressure SEM inside the specimen chamber were investigated using Monte Carlo simulation. In order to improve the signal to noise ratio for the electron detection, we suggest to use helium gas-based mixture. The Helium gas is well known to reduce the skirt effect due to its low elastic scattering cross-section. The addition of an ionizing gas such as hydrogen or nitrogen is proposed to increase the inelastic scattering cross section which is mainly responsible for the ionisation process taking place during the beam-gas interactions. For all the mixtures (except He–Argon), the main results show that the skirt is slightly modified with the increase of the pressure. For the BSE detection, the signal to noise can remain high and gives a good contrast in imaging. Moreover, the presence of an ionizing gas will favour the ionizing process which is very important in beam-based electron detection. In this case, an increase of the signal to noise ratio can be expected.

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1. Introduction

The high-pressure scanning electron microscope (HPSEM) can be defined as a SEM which allows the examination of specimens in a gaseous environment, operating at pressures up to 300 Pa. The high vacuum in the electron-optical column is separated from the high-pressure zone in the specimen chamber by differential pumping through a pressure-limiting aperture (PLA). The electron beam initially propagates unhindered through the electron optics until it approaches the final aperture. Between this aperture and the specimen surface, the number of electrons scattered from the beam increases exponentially with increase in distance traversed in any environment (high or low pressure) but the magnitude of this scattering can be very different. Many instrumental parameters are directly related to the skirt and are discussed by different authors [1–5]. The average number of collision per electron, as defined by Danilatos [2] is given by:

\[ m = \sigma_{PD}/kT \]  \hspace{1cm} (1)

where \( \sigma \) is the total scattering cross-section which varies with the electron energy, \( T \) the absolute temperature of the gas, \( k \) the Boltzmann constant, \( P \) the pressure and \( D \) the working distance, which is the distance between the last pressure-limiting aperture plane and the plane of the electron impact. Consequently, low spreading corresponds to low \( m \) values. From Eq. (1), by lowering the cross-section, the pressure or the working distance we will reduce \( m \). The issue of reducing the beam spreading has been discussed by several authors [1,3,4,6]. Lowering the gas pressure in the specimen chamber reduces the density of gas atoms or molecules, which in turn reduces the \( m \) value. Another way is to raise the accelerating voltage and hence the electron energy, since this lowers the cross-section. In a previous work [7], we have shown that if the beam radius varies between 1 and 100 nm, the resolving power of the HPSEM seems to be independent of the beam-gas interactions, which are in turn independent of the beam width and we have also concluded that for imaging with a low probe diameter in the high-pressure field emission SEM, helium can be recommended. However, owing to the high ionisation energy of helium gas, there is a low production of positive ions which can induce several problems as charging effect for the imaging. In this paper, we consider the choice of helium gas-based mixtures in the specimen chamber. Indeed, in order to increase the positive ions production, we suggest to add a quantity of other gases (hydrogen, water vapour, nitrogen...) which might facilitate the ionisation process. Another way for limiting the charging phenomena is to reduce the accelerating voltage as in conventional SEM. This, in addition, contributes to increase the collision probability of the electron with the gas particles (atoms or molecules) and hence, the gas ionisation efficiency with however, a concomitant increase of...
the electron beam skirt. The main purpose of this paper is thus to investigate the influence of helium gas-based mixture on the electron beam parameters in the low vacuum zone of the SEM with low beam energy conditions. In particular, estimates are given on the change of the beam profiles resulting from the scattering of the primary electron beam and the skirt radii of the electron beam are evaluated for each mixture. The influence of such mixture on electron detection is discussed.

2. Theoretical background

2.1. Monte Carlo simulation

In the regime of the collision called single scattering, of the incident electron beam and the surrounding gas molecules, the majority of the electrons are undergoing about one scattering event. A single scattering model has been previously developed [8,9] which can provide predictions about the radial intensity distribution of the scattered electrons, the fraction of unscattered particles (collision with a probability \( p \)), the mean free path \( \lambda \) for a random number \( n \), the total cross section \( \sigma \), of the gas viscosity \( \nu \), the absolute temperature \( T \) and the gas molar mass \( M \).

\[ \sigma_{el}(E) = \sigma_{el}(E) + \sigma_{in}(E), \]
\[ p_1 = \frac{\sigma_1(E)}{\sigma_{el}(E) + \sigma_{in}(E)}, \]
\[ p_e = \frac{\sigma_e(E)}{\sigma_{el}(E) + \sigma_{in}(E)}, \]
\[ p_i = \frac{\sigma_i(E)}{\sigma_{el}(E) + \sigma_{in}(E)}, \]

where \( p_1 \) and \( p_e \) are respectively the elastic and inelastic fractional probability. The sum of the fractional probabilities is equal to the unity and the interval [0,1] is divided into segments of length proportional to these probabilities. The kind of collision is determined by the value of a third random number which falls either into the inelastic or elastic part.

The actual distance that an electron travels between successive scatterings will vary in a random way. To account for this fact in the Monte Carlo simulation, the step length between two collisions is written as: \( \text{Step} = -L_e \ln(RND) \) cm.

Here, \( L_e \) is the elastic or inelastic electron mean free path.

As the electron travels through the gas, considered as a dilute solid, its energy decreases and since the scattering event is energy dependent, the instantaneous energy at any time needs to be calculated. The energy loss, denoted by \( dE/ds \) could be given by the Bethe relation as [11]:

\[ \frac{dE}{ds} = -78.500 \frac{\rho Z}{A_w E} \log \left( \frac{1.166E}{J} \right) \text{keV/cm} \] (2)

where \( E \) is the energy of the electron (in keV), \( Z \) and \( A_w \) are, respectively, the atomic number and atomic weight of the target (here the gas), \( \rho \) is the density of the target, and \( S \) is the distance measured along the electron trajectory, \( J \), which has units of keV, is the mean ionisation potential, that represents the average rate of energy transfer due to all possible inelastic events (i.e. the excitation of X-rays, Auger electron, phonons etc.).

However, at relative low energy, Joy and Luo [12], rewrote the energy loss equation (2) by incorporating an energy dependent mean ionization potential \( J^* \) instead of the constant \( J \). This modified potential is given by \( J^* = \frac{1.166E}{J} \). By replacing the latter expression of \( J^* \) into (2), one obtains:

\[ \frac{dE}{ds} = -78.500 \frac{\rho Z}{A_w E} \log \left( \frac{1.166E}{J} + 0.99 \right) \]

which can be taken approximately as:

\[ \frac{dE}{ds} = -78.500 \frac{\rho Z}{A_w E} \log \left( \frac{1.166E}{J} + 1 \right) \text{keV/cm} \]

As demonstrated by the authors, this expression is accurate even at energies of 100 eV or below, and also avoids the difficulty in Eq. (2) that cannot be evaluated for \( E < (J/1.166) \). Under both elastic and inelastic scattering events, the energy loss \( \Delta E \) along the step could be calculated using either Eq. (2) or (3) as: \( \Delta E = \text{step}(dE/ds) \).

For elastic event, the new direction after each kind of collision is determined by the angular differential form of the Rutherford cross-section, which yield [13]:

\[ \cos(\theta) = 1 - 2aRND/(1 + a - RND) \]

In this equation that can generate a unique scattering angle \( \theta \) in the range of [0, 180°]. \( RND \) is a random number drawn automatically by computer, and \( a \), a scattering factor [14]:

\[ a = 0.00342 Z^{0.87}/E^2. \]

Similarly, the azimuthal scattering angle could be given randomly by:

\[ \varphi = 2\pi RND \]

For inelastic scattering event, the inelastic scattering angle \( \theta_i \) is derived from [15]:

\[ \theta_i = \Delta E/E \]
2.2. The scattering cross section

An analytical derivation of the differential cross-section has been presented by Lenz [16] and adapted by Jost and Kessler [17] and Danilatos [2]. The differential cross-section is the sum of elastic and inelastic terms. For atoms with atomic number Z; the elastic term is:

$$\frac{d\sigma}{d\Omega} = \frac{A\pi}{16} \left[ \sin^2 \left( \frac{\theta}{2} \right) + \sin^2 \left( \frac{\theta_0}{2} \right) \right]^2$$

(4)

and the inelastic term

$$\frac{d\sigma}{d\Omega} = \frac{A(\theta^2 + \theta_0^2 + 2\theta_0^2)}{\left(\theta^2 + \theta_0^2 + \theta_0^2 + \theta_0^2\right)^2}$$

(5)

where

$$A = \frac{\lambda^4 Z^2 (1 + \frac{E_0}{mE})^2}{4\pi^2 a_0^2} \theta_0 = \frac{\lambda}{2\pi R} \theta_0 = \frac{J}{4E}$$

(6)

with \(\lambda\) the electron wavelength given by:

$$\lambda = 1.226 \times 10^{-9} [E(1 + 0.9778 \times 10^{-6}E)]^{\frac{1}{2}}$$

\(E\) the energy of electron beam in eV, \(R\) the atom radius, \(J\) the ionisation energy of the gas in eV, \(E_0 = 511 \times 10^3\) eV the rest electron energy, \(a_0 = 5.29 \times 10^{-11}\) m is the Bohr radius.

The atom radius may be derived from \(R = \frac{|f_e(0)|a_0}{Z^2}\) where \(f_e(0)\) is the scattering amplitude for electrons [1,18].

In the case of molecular gases, the total scattering cross-section for a molecule is not simply the sum of the elastic and inelastic cross sections. Additional effects are present due to the binding of atoms in molecules. In order to calculate the molecular cross-section, we use the theory developed by Danilatos [1]. For the many scattering centres formed by the atoms in a molecule, let the minimum distance between centres be \(d_0\) and the maximum range of interaction of the incident electron be \(r_m\). In the case of coherent scattering and, if \(\frac{2a_0}{r_m} \gg 1\) and \(\theta_0 \gg r_m\) the differential elastic cross-section of a molecule is given by:

$$\frac{d\sigma_e}{d\Omega} = \sum_n \sum_j f_n(\theta)f_j(\theta) \frac{\sin (s \times r_m)}{s \times r_m}$$

(7)

where \(s = 4\pi a_0^2 \frac{\sin (\frac{\theta}{2})}{\theta}\) and \(r_{nj}\) is the distance between atoms \(n\) and \(j\); \(f_n(\theta)\) is the scattering amplitude, from which we get:

$$f_n(\theta) = \frac{d\sigma_e}{d\Omega}$$

(8)

Thus we have an expression for \(f_n(\theta)\) using Eq. (4).

The case of the inelastic cross-sections can be considered under incoherent scattering and the differential inelastic cross-section is the sum of the individual atom cross-sections as a first approximation.

$$\frac{d\sigma_i}{d\Omega} = \sum_j \frac{d\sigma_{ij}}{d\Omega}$$

(over all \(j\) atoms)

3. Results and discussion

3.1. Calculated cross-section for the different gases

The elastic and inelastic scattering cross-sections calculated at 5 kV for H$_2$O gas are plotted in Fig. 1. Our results and those of previous workers [2,6] show that the two scattering processes result in very different angular distributions. Elastically scattered electrons are deflected through angles larger than 10$^{-2}$ rad. The relative higher angle for elastic events determines the width and magnitude of the large scattered region which surrounds the beam. The inelastic electrons are scattered through small angles, as low as 10$^{-4}$ rad, and it is the inelastic events that determine the form of the beam profile close to the axis.

By numerical integration of the sum of Eqs. (4) and (5) for atomic gases and, respectively, Eqs. (7) and (8) for molecular gases, the total cross-section of Ar, He, H$_2$, O$_2$, N$_2$, H$_2$O and Air have been calculated and plotted vs. beam energy in Fig. 2A.

The calculated total scattering cross sections compared with those used by Danilatos [1] are smaller. This result is attributed first to the choice of the \(J\) value in equation (6). The same atom or molecule may be submitted to successive ionisations by the electron beam. Thus we have chosen the mean ionisation potential instead of the first ionisation potential as suggested by Reimer [19].

In the field of low energy, our calculated cross section has been compared with experimental results for water molecule [20], and a good agreement was obtained, which validates the choice of the mean ionisation potential. It must be highlighted that the right choice of \(J\) is of paramount importance since the \(J\) value induces a major change of the cross section at small scattering angles.

The curve for molecular nitrogen is below that of argon and above the curve of water and helium. This comparison shows the straightforward relationship between the total scattering cross section and the mean atomic number. The elastic and inelastic scattering cross-sections are plotted respectively in Figs. 2B and 2C. It is interesting to notice that the elastic scattering for H$_2$O is greater than for N$_2$, and the Ar inelastic scattering cross-section is smaller than for N$_2$. These facts indicate that the previous rule concerning the total scattering cross section and the mean atomic number is not valid for the elastic and inelastic scattering cross sections considered separately.

In order to limit the skirt effect which is very prejudicial to the X-ray microanalysis [21,22] with a loss of spatial resolution due to the beam broadening, it is important to choose a gas with low elastic scattering cross section as helium. However, the helium ionisation energy (24.5 eV) is high comparatively to water vapour (12.35 eV) or nitrogen (13.61 eV). For the imaging aspect, the efficiency of the surface neutralisation is not favoured by only the use of helium gas.

In order to improve the number of environmental electrons and ions which result from the collision between the primary beam and the gas, it is interesting to use a gas with a high inelastic scattering cross section like N$_2$. Indeed, it is well known in the solid, that during the beam bombardment, it is the inelastic collisions that are mainly
responsible for X-ray emission and ionisation of the atoms of solid. In the gas which in our model, is assimilated to a dilute solid, the same phenomenon can take place and the inelastic interaction is the only process to give element to the ability of the gas ionisation which is very important for the imaging in the high pressure SEM. Fig. 2C shows clearly that helium possesses the lowest inelastic scattering cross section comparatively to Ar, H₂O and N₂. It is why we proposed to study the influence of the helium gas based mixture on the electron beam parameters in the low vacuum zone of the SEM with low beam energy conditions.

3.2. Electron beam

A Monte Carlo code was used to simulate the electron path in the specimen chamber and to calculate the electron beam skirt, with accelerating voltage of 5 kV and pressures of 10 and 100 Pa respectively. The working distance was taken equal to 10 mm, which corresponds to the analytical distance for X-ray microanalysis in numerous SEM. Up to 10⁵ electron trajectories were computed for each gas mixture. The nature of the gas varied from gases usually used in high pressure SEM as He gas, water vapour and Air gas to the helium-based mixtures as He–Ar, He–O₂, He–N₂, He–H₂O and He–H₂. In each case, during the calculations, the gas mixture was assumed to be composed of 90% of He. In order to quantify the beam spreading, \( r_{0.9} \) is defined as the radius in the plane of the electron impact that contains 90% of the scattered electrons [1,6]. The unscattered fraction belonging to the primary beam is first subtracted from the total current and it is the remaining component that is characterized by the broadening measured by the \( r_{0.9} \) radius. To determine the profile, we extend this definition to different fractions of scattered electrons.

Fig. 3A shows the fraction of scattered beam obtained versus the gas nature at 10 Pa. In all cases, the shape of the curve is composed of inner and outer regions that correspond respectively to the inelastic and elastic scattering. The outer region can vary from micrometer for helium gas to several millimetres under air gas. This fact confirms that the skirting effect is directly related to the elastic cross section. When the pressure is raised to 100 Pa (Fig. 3B), as expected, the fraction of the scattered beam is more important for each gas. These results confirm all previously published results that helium reduces drastically the skirt effect which is very limiting for a good practice of X-ray microanalysis [21,23]. In fact, helium allowed stability of the results of microanalysis and improvement of the chemical contrast of images. The increase of the signal-to-

![Fig. 2.](image_url)
noise ratio allows an improvement of the detection limit of the microanalysis system when helium is used.

For the imaging aspect, in order to favour the ionisation, another gas with a high inelastic cross section is added. Under helium gas mixture (Figs. 3C and 3D) at 10 and 100 Pa, it appears in all cases that the fraction of scattered beam follows the helium behaviour. Moreover, the extent of the outer region effect is little changed except for the argon case because the argon elastic scattering cross section is very important comparatively to the Helium. It is interesting to notice that the behaviour for the He–H₂ mixture is very similar to that of helium gas. For the other gases (N₂, H₂O, O₂), an intermediate behaviour is observed which is linked to the elastic cross section value of each gas. An interesting feature is the fact that the increase of the scattered fraction close to the axis is slightly modified. This means that the unscattered part which gives the useful signal for the electron detection will be little disturbed. In the electron detector based on the beam gas interactions such as the gaseous secondary electron detector (GSED), specimen current and the photoscintillator detector, a positive effect due to the ionisation can be expected.

3.3. Skirt radius

In order to compare the influence of the gas environment, we have plotted the skirt radius $r_{0.9}$ vs. pressure (Figs. 4A and 4B). It appears that for He, the variation of the skirt radius does not depend on the pressure in the investigated range from 1 to 100 Pa at 5 kV. It is interesting to note that the skirt radius increases slowly with the increases of the pressure for the other mixtures. The only exception concerns the He–Argon mixture due to the important effect of the elastic scattering cross section of argon. The small change of the skirt radius is in agreement with the beneficial effect with the use of helium gas in BSE detection which has been shown by Oho et al [24] and Belkorissat et al [7]. In these papers, the authors have shown that the signal to noise ratio remains high when using helium even when the pressure inside the specimen chamber is substantial. When the mixtures are compared, the He–H₂ seems obviously to have the same behaviour as pure helium. So, the backscattered electron detection in presence of a helium gas mixture will keep this positive aspect. For the beam-gas interaction based detector (GSED, specimen current and the photoscintillator detector), for the different
helium gas mixtures, the amount of secondary electron emitted at the impact point is due essentially to the unscattered part of the primary electron beam. As the scattered part remains nearly constant, we can expect that the amount of the secondary electron emitted from the solid is slightly modified. However, the presence of an ionizing gas will permit to improve the ionisation process and the generation of environmental secondary electron will be favoured. In this case, the signal to noise ratio will be increased.

4. Conclusion

The effects of different gas environment used in high pressure SEM inside the specimen chamber were investigated using Monte Carlo simulation. In order to improve the signal to noise ratio for the electron detection, we suggest to use helium gas based mixture. The Helium gas is well known to reduce the skirt effect due to its low elastic scattering cross-section. The addition of an ionizing gas such as hydrogen or nitrogen is proposed to increase the inelastic scattering cross section which is mainly responsible for the ionisation process taking place during the beam-gas interactions. For all the mixtures (except He–Argon), the main results show that the skirt is slightly modified with the increase of the pressure. For the BSE detection, the signal to noise can remain high and gives a good contrast in imaging. Moreover, the presence of an ionizing gas will favour the ionizing process which is very important in beam-based electron detection. In addition, the presence of such ionizing gas leads to work at lower pressure, which reduces the beam skirt phenomena. In this case, an increase of the signal to noise ratio can be expected.

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