A review on correction methods to solve limited representativeness in mineralogical analysis of measured liberation data.

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Recent trends in the improvement of the mineral liberation measurements are briefly reviewed. Mineral liberation is the parameter used to suggest action to the flotation process which is intended to render better separation efficiency. The exposed grain surface area of 2D imaging of a polished section with stereological correction is proven to be the appropriate method to measure mineral liberation. Supplementary to 2D imaging, 3D micro CT scanner has also been used to determine the internal structure of the materials to analyze the volumetric grade of mineral of interest. There is a significant development that has been made in correcting measured liberation data acquired from 2 dimensional plane to 3 spatial dimensional. Although progress is being achieved, there are still challenges facing correction methods. Therefore, the advantages and disadvantages of specific issues, such as point spread function and excitation energies, facing the correction methods are discussed.

Keywords: Mineral liberation, 2D imaging, Modelling, 3D data

1.Introduction

The aim of this review is to highlight the sources of improvements and limitations of mineral liberation measurements in two-dimensional (2D) imaging information and stereological correction methods which are used to transform 2D mineral liberation information to three-dimensional (3D) information. Lastly, the recommendations are made based on a recent acquired knowledge.

So far, mineral liberation data are often obtained from 2D image which is acquired from scanning electron microscopy (SEM) with energy dispersive X-ray (EDX) attached to it. The equipment can reveal more information about the sample such as mineral crystallography, external morphology (texture), topography and chemical composition composed in the sample. However, it cannot display the full orientation of particles in all different directions, this causes the limitation in quantifying the mineral liberation of a particle in three-dimensional space. Though SEM/EDX has features that can be used to manually measure the two-dimensional mineral liberation of cross-sectioned surfaces, but they are biased because they are operator-dependent and not reproducible. Nevertheless, the information of mineral grade and volume grade provided by EDX is useful in calculating mineral liberation. Yet, calculated mineral liberation results have been found to overestimate and often underestimate the true liberation of ore particles (Wang et al., 2018; Ueda et al., 2016). However, the perimeter grade and volume grade data generated still hold valuable information set, which

usually used to transform two-dimensional to three-dimensional data. The mineral liberation measurements have been found to be function of the particle orientation with respect to mineral phases present including shape, size and mineral texture (Hilden and Powell, 2017; Wang et al., 2018; Mariano et al., 2016; Zhang and Subasinghe, 2013a; Lätti and Adair, 2001). All these attributes are dependent on each other to render the true mineral liberation. As much as grade volume can be measured, the true three- dimensional volume grit (voxel) of particles are not necessarily accounted for. Consequently, for it to be incorporated in the 3D measurements, it must be estimated or assumed which could compromise calculations to represent true voxel.

To resolve the abovementioned 2D image challenge, different stereological techniques have been employed to correct mineral liberation measurements. The methods are aimed at eliminating or minimising the 2D bias information. Yet, some assumptions and estimation of unknown parameters are made in most of the correction method which also introduce some degree of bias information. These assumptions are brought about by the unknown attributes especially unpredicted particles orientation that are undetected from the surface of the polished section. It has been noted that stereological methods have proved useful in reducing the problem of 2D liberation projection of 3D liberation data (Ueda et al., 2017) . Nevertheless, stereology is supposed to be unbiased (Brown, 2017). Though rigorous sampling has been used to eliminate discrepancies of many correction methods, but the resultant data still contains some degree of bias due to variable assumptions and estimation made in the correction methods. Supplementary, the filtering efficiencies incorporated in some methods may omit some important liberation information or exaggerate the final measurement results since the expected three-dimensional particles are unknown.

The other method that is rarely used but recently, found more attention to measure mineral liberation is X-ray computed tomography. The 3D data set is projected by group of 2D slice images obtained from microCT scanner. Though it can show the tomography of particles in different directions, it is still having limitations when it comes to quantification of mineral liberation measurements. Therefore, some employ software that can quantify the liberation which usually work for coarse particle sizes. As much as this technique has been continuously developing, it still has limitations for accurately measuring mineral liberation at fine particle sizes. This is due to its inability to separate fine particles from each other, which makes it difficult to measure liberation at that sizes. This is also due to challenges in resolution. As such, many mineral processing industries make use of SEM based images instead, to analyse liberation degree.

When SEM based imaging is developed generally the point spread function, energy excitation of atoms, monochromatic assumptions of X-ray beam and high resolution play a big role in generating a high-quality image. Any assumptions or compromises made during image development specifically of the latter attributes could affect information of image acquisition from which a 2D mineral liberation data will be extracted. Therefore, when correction methods are employed, they will inherit assumptions and compromises brought by the procedure followed during image development. Supplementary to that, the 2D data above-mentioned challenges could also be included. Lastly but not least, the assumptions or estimation that come with correction methods could also increase the total number of assumptions. This hierarchal addition of assumptions and estimation introduced, starting from image development to 3D information, are source of bias to the data and the degree of

deviation of final results which supposed to give reflection of true mineral liberation of particles. Thus, bias in this context of study can be simply defined as the difference between true mineral liberation of particles and calculated mineral liberation of particles. Unfortunately, in mineral processing the true mineral liberation is generally unknown, hence a need of having unbiased data is critical. This is owed to the complexity and unpredictability of particles geometry which makes it difficult to accurately predict true mineral liberation orientation.

Since the imaging of SEM/EDX technique is most widely used for the inspection of mineral liberation in mineral processing industry, there is a considerable interest in the understanding of image generation process of the technique from which liberation phenomenon is ultimately measured.

2. Image development and acquisition

It is of importance to understand the advantages and limitations that come with the SEM/EDX technique in the context of image forming which could eventually affect the mineral liberation measurements downstream. The technique uses three detectors namely, (1) secondary electron detector which mostly is Everhart-Thornley detector and is used for provision of sample surface topography, (2) backscattered electron (BSE) detector to render an image with elements distribution by using atomic number contrasts and (3) X-ray detector which is frequently energy-dispersive X-ray spectroscopy is for identifying elements present by making use of the contrasted elements distribution from backscattered electron image. The BSE imaging information with conjunction with the X-ray analysis are used for composition quantification and mapping of their distributions. The different types of electrons and X-rays are emitted upon the contact between primary beam and the sample, as it is shown in **Figure 2**. These different types are emitted according to the penetration depth of the incoming excitation energies.

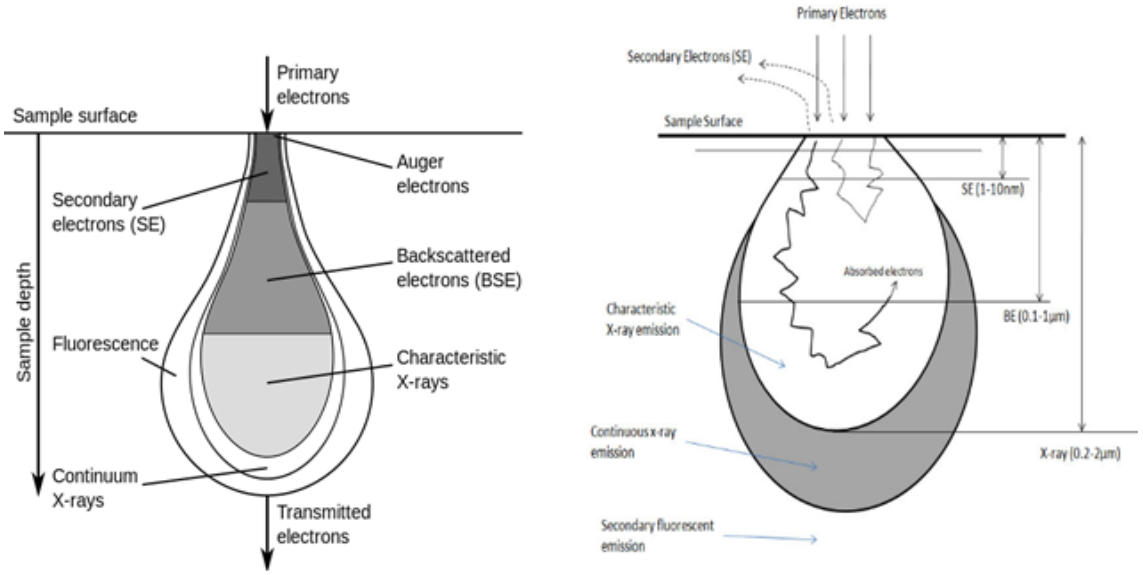

Figure 2: Shows the different types of electrons and X-rays emitted upon the contact between primary beam and the sample. The depth level required are also indicated.

The 2D mineral liberation measurements require high-quality SEM imaging which is frequently brought by high spatial resolution. The spatial resolution is also known as point spread function.

2.1 Point spread function

Point spread function can simply be defined as blurring of object image as shown in **Figure 3.** With the years progressing, knowledge has been established of how point spread function differs from one system to another. In the case of SEM/EDX technique, point spread functions are caused by current density from electron beam, overvoltage, and elemental density and signal depth from polished section. These PSF research began from as far as the inception of image intensifier and CCD cameras. Though, it was noticed that CCD cameras are resolution limited by their 8 bits, image intensifiers demonstrated the potential of providing higher resolution (Kakumanu, 1994).

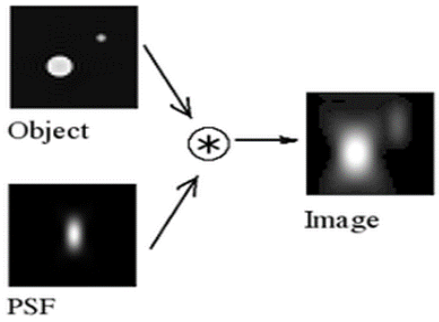

Figure 3: Demonstration of the point spread function in an image development.

Although there has been extensive work on examining and optimizing the PSF in two-dimensions, there is still less known about the true PSF along the Z- axis of thick sample of polished section. In addition, the spatial resolution of the image from SEM- integrated MLA's is dependent on the number of lenses used. The more the lenses the higher the image quality.

2.2 Excitation energies

The voltage is often accelerated to emit X-ray characteristic from within the sample and it is associated with excitation energy. As a result, more information about elemental identification can be extracted and be represented as a spectrum from which area grade and mineral volume are obtained. Eventually, mineral liberation measurements.

The interaction estimation between X-ray and sample have been calculated using a relationship between specimen density, accelerating voltage and critical excitation energy as it is seen in equation 1.

$$\mathbf{R = 0.064\ (E_0^{1.68} - E_C^{1.68})\ /\ \rho} \quad \text{.....(1)}$$

where:

R = spatial resolution in um;

Eo = accelerating voltage in keV;

Ec = critical excitation energy in keV;

ρ = mean specimen density in g/cc.

For the generation of characteristics X-rays, the excitation energy, in the form of voltage, should be high enough to generate sufficient X-rays and low enough not to generate continuum X-rays which are considered to minimise the visibility of the characteristics X-rays of the composition spectrum (Hafner, 2006). Therefore, if the excitation energies are highly accelerated by accelerating the voltage, the spatial resolution is minimised so is the number of characteristics X-rays emitted. In turn, the image quality and information carried could be biased. In the same breath, if the excitation energy is not sufficient some elements could be missed. This concept is demonstrated by Beer's law.

$I/Io = \exp(-\mu_M \rho d)$ …….. (2)

where:

I/Io = fraction of X-rays transmitted;
d = thickness;
ρ = material density;
μ_m = mass absorption coefficient

Somehow, the cut off limit of voltage still need to be applied, to provide enough qualitative information.

Some SEM techniques, at times once the qualitative analysis of EDX is acquired, the user uses his knowledge of chemical principles and ore sample under study to physically confirm the results. Furthermore, if the user does not confirm that number of counts are statistically sizeable, other peaks might be missed or appear as absent. In the SEM operation if the acquisition time is small, some peaks are lost in the background, see the figure below (Hafner, 2006). Consequently, all of these could also add results bias which contributes to 2D mineral liberation data. Thus, eventually stereological measurements.

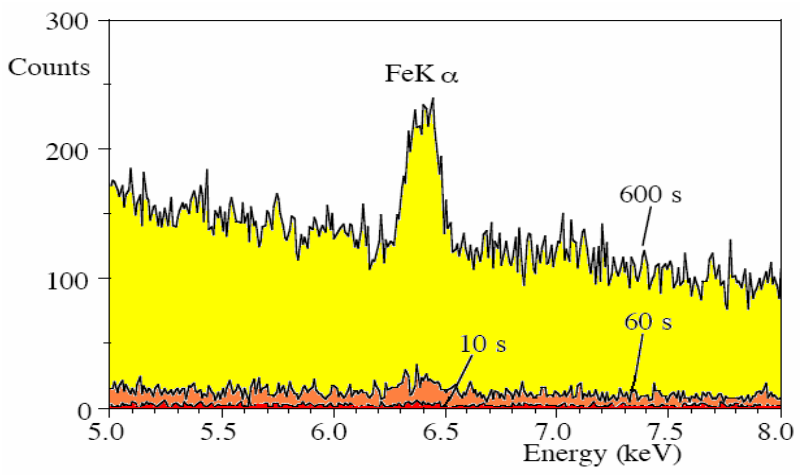

Figure 4 displays the peaks which could be lost in the background in the event of small acquisition time (picture by Hafner, 2006).

Other SEM techniques have incorporated 'Noran System Six software" NSS automatic qualitative analysis, to overcome the latter user limitations.

However, in the SEM imaging chain, there have been challenges of determining accurate point spread functions which are brought by electron beam current density, overvoltage, specimen density, signal depth from within the sample and the low resolution of backscattered electrons of which all contribute to compromised information signals and imaging if not measured well.

As much as there have been attempts to accurately measure all the parameters mentioned above to improve the quality of the SEM image, there are still some improvements required in the methods published. For example, the depth-resolution has been perceived as a function of 2D probe shape and there is little information known about the probe shape along the optical axis.

Supplementary to the challenges and improvement made thus far, the scanning electron microscopy with energy dispersive X-ray incorporated, has also been developed to enhance the imaging of material ores to the atomic level. One of the recent additions to improve spatial resolution/ point spread function of the image is making use of cathodoluminescence detector (Gustafsson and Kapon, 1998). Given all the improvements mentioned above, however, some of the 2D measurements difficulties are still under investigation. Therefore, the true orientation still an unresolved issue. Consequently, many researchers paid more attention to stereology science. Many approaches use stereological correction to estimate what could transpire in 3D mineral liberation orientation.

3. Some of stereological correction methods used thus far.

3.1 Development in measuring mineral liberation in three-dimensional space

In this review, the one- and two- dimensional mineral liberation measurement models are not covered as extensive research have been conducted and well published. Much work has been developed on two modes which began since Gaudin (1939) and other researchers continued to improve the prediction of mineral liberation measurements by building on the established methods (Petruk, 1978). For example, literature shows how many researchers such as Barbery, Evans, Ueda and their colleagues (Mariano, 2016; Ueda et al., 2017, 2016; Evans et al., 2013) extended Gaudin texture model. Whilst others applied different models with different functions and coefficients (King, 2000; Steven Spencer; David Sutherland, 2000; and many more).

In 2013, Zhang and Subasinghe modified Barbery liberation model developed in 1991, to predict liberation in 3D. The modification included the use of probabilistic functions measured using image analysis instead of using ore texture and particle structure assumptions (Zhang and Subasinghe, 2013b). However, they did not factor stereological correction to incorporate the inherent one- dimensional geometrical measurements bias. Therefore, this review focused more on the stereological correction method of two-dimensional mineral liberation data from polished section and artificial three-dimensional particles used to validate or invalidate the correction method in use.

It could be seen from **Table 1** that so far in mineral characterization, stereology has been the mostly used estimation technique to quantify two-dimensionally acquired mineral liberation measurements. Most models are a list of equations which depend on several assumptions regarding the particle internal structure, size of mineral phase, liberation of the phases, volume fraction, and binary phase of the particle, shape, area grade, volume grade, and many

more. Therefore, the 2D measured values would be transformed to 3D using some of equations mentioned below. Thus, as much as progress has been made in transforming 2D liberation distribution/ data into 3D, all stereological methods used so far are deficient of a quantitative basis. As a result, this deficiency brought about researchers designing artificial particles already in 3D space with known internal structure and characterizing them to overcome the quantification. See **Table 1**.

The artificial 3D models are also applied to examine the validity of these assumptions and determine the sensitivity of stereological correction methods used to measure true mineral liberations and their deviations from such assumptions. This is performed by sectioning the created 3D particle into 2D image and thereafter conduct quantification of stereological bias (Ueda et al., 2018, 2017, 2016). The **Table 1** below, shows some of the equations of such of liberation models.

Table 1: Stereological correction calculated by characterising 3D data to obtain 2D.

References	**Stereological Correction 3D – 2D**
Wang et al. (2018) -Same sample analysed directly from 2D MLA and 3D-HRXMT, separately.	$Stereological\ correction\ factor = \frac{100 - [percentage\ of\ 0\%\ class\ by\ 3D\ HRXMT]}{100 - [\ percentage\ of\ 0\%\ class\ by\ MLA]}$ *where 0% class represents 0% grade class ('liberated' gangue minerals), and 3D HRXMT represents for 3D High Resolution X – ray MicroTomography whilst MLA stands for Mineral Liberation Analyser.* *Here, exposed grain surface areas obtained from 3D HRXMT and MLA were compared and stereological correction factor was then calculated as per equation above.*
Ueda et al. (2018) -3D-particles were modelled	*Stereological bias assessment (for phase A)* $= L_A^{2D-3D} = L_A^{2D} - L_A^{3D}$ $(for\ phase\ B) = L_B^{2D-3D} = L_B^{2D} - L_B^{3D}$ *where,* L_A^{2D} *and* L_B^{2D} *represents two – dimensional liberation of phase A and B, respectively whilst,* L_B^{3D} *and* L_B^{3D} *denotes three – dimensional liberation of phase A and phase B, respectively.*
Ueda et al. (2017) -3D-particles were modelled	*Stereological correction*: $\wedge_A^{dif} = \wedge_A^{2D} - \wedge_A^{3D}$ ..... (1) *Then, equation (1) was substituted in equation (2) as it is shown below,* $\wedge_A^{3D'} = \wedge_A^{2D} - \wedge_A^{dif}$ ..... (2) *where* $\wedge_A^{2D}$ *and* $\wedge_A^{3D}$ *represent two – and three – dimensional liberation distribution parameters, and* $\wedge_A^{3D'}$ *denotes stereological correction.*
Ueda et al. (2016) -3D-particles were modelled and simulated	*Stereological bias quantitative assessment*: $(for\ phase\ A) = L_A^{2D-3D} = L_A^{2D} - L_A^{3D}$ ..... (1) $(for\ phase\ B) = L_B^{2D-3D} = L_B^{2D} - L_B^{3D}$ ..... (2) $\sigma_A = \frac{L_A^{2D-3D}}{L_A^{2D}}$ ..... (3) $\sigma_B = \frac{L_B^{2D-3D}}{L_B^{2D}}$ ..... (4) *where,* L_A^{2D} *and* L_B^{2D} *represents two – dimensional liberation of phase A and B, respectively whilst,* L_B^{3D} *and* L_B^{3D} *denotes three – dimensional liberation of phase A and phase B, respectively.* σ_A *and* σ_B *are defined as the fraction of the overestimated liberation degree.* *This stereological bias quantitative assessment was calculated to examine the effect of sampling number on mineral liberation.*

As much as their quantification procedure to quantify the overall characterisation of stereological bias by using numerical analytical approach has presented positive data to assess stereological bias. It is worth noting to mention that since their method do not make use of SEM/EDX imaging, the assumptions inherent from the equipment are eliminated. However, further work is still to be done for real ore particles since the binary particles used in their study were artificial. As a result, this pose a limitation for practical use in mineral processing.

The designing of 3D particles to overcome stereological bias begun as far as 1982 by Serra, and other researchers were (Gay, 2004, 1999; Ueda et al., 2017;). These 3D characterization methods directly quantify 3D particles as opposed to estimated quantifications. As a result, due to the new data acquisition from direct 3D quantification, there are new improvement surfacing and so is the development of new comprehensive software's for geometrical analysis for mineral liberation. In addition, the ability to perform direct 3D characterization, alignments of other models can be conducted and so is the correction of other assumptions made during stereological correction.

4. Conclusions

With SEM/EDX imaging, as much as X-rays can be generated from the sample as deep as 2µm, there are depth limitations due to X-ray absorption, continuous X-ray generation that form most of the spectral background as the beam progress deep in the sample, which makes it difficult to differentiate the minor and trace elements in the system. Therefore, the mineral liberation measurements corresponding to the data obtained from such system might be misleading. The thickness of the sample also plays an important role in SEM- based image since electrons cannot penetrate through as compared to electrons from transmission electron microscropy.

Different models have been illustrated to predict the laws which govern true mineral liberation orientation. Those models have been probed with different parameters (input signals) and systems responses have been observed, analysed and compared to. Notably, there are relationship of some parameters realized through modelling. Many properties contributing to liberation orientation have been discovered. Though many researchers predominantly use texture models for liberation calculations, combination of varying parameters as a function of texture are used and so is the application of different stereological correction methods and assumptions of some parameters. Hence, there is not one texture model realized for liberation estimation. Consequently, since models are not reality instead trying to emulate the true systems, it is noted that all models are performed by stimulating them with input signals and they only depict the aspect of what they have been stimulated with. As a result, it is noted that to this point in mineral liberation measurement, no one standardised method is applied to calculate or measure liberation orientation of real particle system. However, the true mineral orientation of the real ore particles still an unresolved issue. Therefore, there is still further research to be done in this area.

Since 2D mineral liberation information have no direct correspondence with 3D mineral liberation structure. Different researchers attempted to associate the 2D information to 3D mineral liberation structure by using different correction factors to cater attributes lacking in 2D. The methods also included rigorous sampling techniques, statistical theories and stochastic geometry. However, not all of these tools can be applied to one/every correction

method developed for mineral liberation calculation. This could be because most correction method are assumption-based than design based stereology.

In this work, we have reviewed the recent studies in which it is observed that particles can take any orientation, shape or size in the z- direction, and there is no known true mineral liberation in mineral characterisation, one correction method might deviate from the truth whilst the other might be close to the truth. Meaning, some method might be intermittent based on the particle population given at any point.

Therefore, to summarise the literature compiled above, it seems that there is no one standard stereological correction factor recognised industry-wide for mineral liberation measurements. So far, vast knowledge about orientation of the particles can be approximated as a function of mineral liberation based on the latest findings of quantification procedure developed from characterised 3D particles. Some boundaries can be established and some derivatives are now known exactly. There are great improvements since Gaudin, 1939. The errors have been minimised but more work is still to be done with real ore particles.

5. Recommendations

One of the solutions to overcome the challenges faced by 2D data in addition to stereological corrections for mineral liberation is continuous upgrading of imaging techniques where depth resolution of the thick samples could be increased or penetrate through the section to expose sections from within the samples. Thus, resulting in the images representing all geometrical view of particles with pronounced contrasts of each phase present. These might correct some of the assumptions made so far, to estimate true liberation. Furthermore, if we could find a way of imaging 3D real ore particles, or even 4D imaging, without limitations many distortions, estimations and assumptions will fall off. The other solution is the encouragement of more development of mineral liberation models with many measured variables. To overcome the agglomeration of fine particles, the resolution properties of X-ray computed technology might need to be improved. So is the sample preparation of fine particles for the equipment. Designed based stereology could likely yield results that are close to true mineral liberation unlike assumption based stereology.

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