

Characterisation of Particulate Matter Using Computer Controlled Scanning Electron Microscopy

Abstract

Scanning electron microscopy with X-ray analysis provides a high resolution characterisation technique for individual particles. Automation using computer control for data acquisition and manipulation offers important advantages of speed and objectivity. One such method is demonstrated with discussion of its strengths and weaknesses. Particle imaging and selection are described, followed by discussion of acquisition of shape and chemical information, statistical examination, presentation (plotting and tabulation) and classification. Examples are drawn from previous applications in other fields but the potential of the technique in the field of archaeology is considered.

INTRODUCTION

Fly ash particles provide an important historical record of the influence of industrial activity on the extent of acidification of the environment (Rose *et al.*, 1990, Rose, this volume). In addition some of these particles are widely believed to add directly to the weathering of building materials, in combination with an acid environment (Del Monte *et al.*, 1981; 1984, Camuffo *et al.*, 1982; 1983). Particles from other natural and anthropogenic sources may also have valuable roles both as tracers of different activities and as possible reagents and catalysts for reactions (Pacyna and Ottar, 1989). It is therefore often valuable to be able to describe these particles in detail and perhaps to classify them in terms of their composition or origin.

A number of techniques exist which give data on the bulk chemistry and/or mineralogy of materials. These analytical techniques may be destructive

(eg atomic absorption spectroscopy (AAS), inductively coupled plasma atomic emission spectroscopy (ICPAES)) or non destructive (X-ray fluorescence (XRF) or X-ray diffraction (XRD) analysis) but all will give an average over a relatively large sample. Chemical analysis in electron beam instruments, on the other hand, allows a high resolution description of the chemical variability across the sample which may add greatly to the bulk analysis data. A trace element may occur in a number of different ways in the specimen, all with different implications. An element which occurs as a trace constituent in every particle in an assemblage will not indicate the same thing as a trace which occurs as a relatively few highly enriched particles and yet both will give the same results in a bulk analysis. If analysis is undertaken in an electron microscope (EM) then there is also unrivalled ability to obtain descriptive morphological and textural information at both high and low magnifications. It is the intention of this paper to introduce the technique of individual particle analysis (IPA) in the scanning electron microscope (SEM), and in particular to consider the opportunities offered by automated systems using computer control of data acquisition. This will not be a review of all systems but will be based on a description of their general capabilities, and will use one particular system for demonstration. A number of extra references are given which give examples of other equipment and its use in relevant studies. Neil Rose (this volume) will present an application where the particular system described here was used to distinguish between carbonaceous fly ash materials from coal and oil burning, and to assess the relative contribution of each to sediment records from a number of lakes.

SAMPLING AND SAMPLE PREPARATION

These vary greatly according to the nature of the material to be studied and the data required. In general particulate material may be deposited directly onto a suitable substrate or may be suspended in a liquid and filtered. Solid samples may be mounted on stubs for examination of surface characteristics or sectioned and polished.

ANALYSIS AN INTRODUCTION TO SCANNING ELECTRON MICROSCOPY

In an electron microscope a beam of electrons is focused onto a specimen and a number of signals result (Fig. 1). These are measured by suitable detectors and a number of possible instrument configurations exist. In the JEOL 733 Superprobe used for this study both morphology (images and micrographs) and chemistry (X-ray data) may be examined. Use is

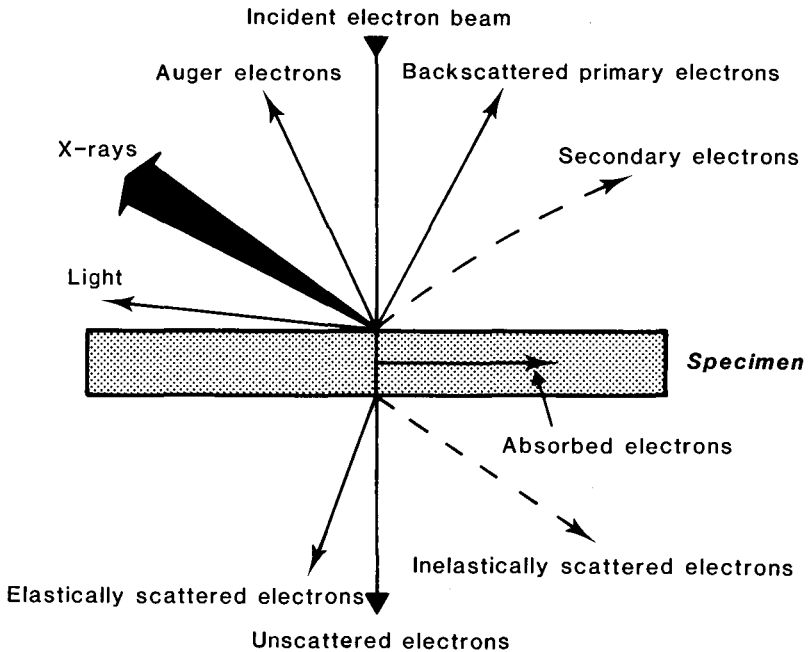


Fig. 1. Specimen/Beam Interactions in the Electron Microscope.

made of three detected signals – secondary electrons, backscattered electrons and X-rays.

This information can be gathered manually by an operator or automatically by a suitably programmed computer. Secondary and backscattered electron images (see below) may be digitised and captured on computer for further manipulation. Converting an analogue signal to a digital one involves splitting the signal up into a number of different levels (termed «grey levels» – 255 levels are used in this system). A grid is superimposed on the SEM field of view and the level is measured at each point on the grid (these points are also known as picture points or «pixels»). The image is built up by displaying the measured value as a tone of grey (hence grey level) on the computer monitor. Thus instead of a smooth waveform variation across an image, there is a series of discrete steps (Normally the resolution of the grid and the monitor it is displayed on is such that an image still appears smooth).

The great advantage that these digital images have is that because each pixel is represented by a number, they can be stored and manipulated by computers. (Perhaps confusingly «grey» levels can be displayed as different colours simply by making the numbers correspond to a shade of that colour rather than a shade of grey).

a. Secondary Electrons

These are electrons that have been ejected from the specimen by the passage of a high energy electron from the beam of the instrument. It is the secondary electron SEM micrographs which resemble optical (light) views of a specimen but with far better resolution, and therefore useful magnification and depth of field, than is available in any other way. They are used to examine texture and morphology in the specimen.

b. Backscattered Electrons

Backscattered electrons (BSE) are high energy electrons from the incident beam of the SEM which have been diverted by a series of collision events in the target (sample). The very useful property that they have is that the output signal strength is proportional to the mean atomic number (also known as the *Z* number). Every element in the periodic table has an atomic number from Hydrogen (the lightest at 1) upwards. Where a particle is a compound of several elements its atomic number corresponds to the mean of the *Z* values for those elements, calculated according to its formula. Thus oxides are lighter than their equivalent metals and have a smaller mean atomic number. The BSE signal strength increases with increase in *Z* value.

This backscatter contrast may therefore be used to perform the equivalent of an on-line density separation — by the technique of BSE thresholding. By selection of suitable upper and lower thresholds features of any given band of mean atomic number can be isolated.

c. X-rays

X-rays are generated as a result of the release of energy which accompanies the movement of an electron from an outer shell to replace a dislodged secondary electron. X-rays can be detected in two ways on the JEOL 733 — by their wavelength or by their energy. The present data were gathered using energy dispersive X-ray spectroscopy (EDS). Each element in the periodic table has a characteristic energy level (or levels) at which it will be detected if present and these are displayed as peaks superimposed on background X-ray radiation. The detector used in this study can measure any element heavier than sodium. The resulting spectra are used to determine what elements are present in the particle as a whole, or at different points on its surface. The X-rays are generated both at and below the surface (cf. Fig. 2) so will reflect what is present throughout this «depth of penetration», usually about 2 microns or less. The height of the peak is proportional to the number of X-ray photons detected and thus the relative proportions of each element present in a feature may be estimated.

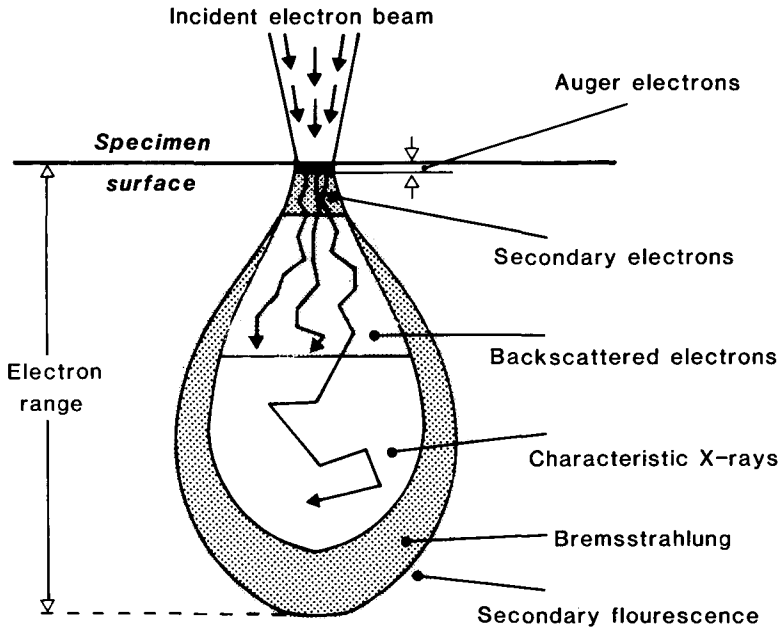


Fig. 2. Depth of Beam Penetration and Signal Emission.

Three different groups of peaks may be seen (termed «K», «L» and «M» series) corresponding to different electron shells in the atomic structure of the element. Not all elements have lines in all series because not all possess the relevant electron shells. The particular combination of number of peaks and energies at which they occur are unique to the element and this is the basis for the ability to distinguish between them. Peaks from different series and different elements overlap in parts of the spectrum, and calculations have to be undertaken to determine which element is present (cf «overlap correction» in the section on automated particle analysis below).

Where appropriate, quantitative X-ray analysis may be undertaken, using chemical standards for calibration (on mounted and polished specimens). In this case a number of corrections are made to the analysis to compensate for atomic number, absorption, fluorescence (Zaf corrections), as well as compensating for overlap and calculation of such things as oxide corrections. In quantitative analysis as many factors as possible are held constant — the sample is sectioned and polished to present a uniform surface in order to keep the geometry of the sample/detector constant. In the analysis of rough surfaces and especially of particulate material this cannot be done and so the resulting analysis is only semi-quantitative. As will be shown, a great deal of useful information can nevertheless be obtained.

X-RAY MAPPING

The distribution of different elements in an SEM field of view may be displayed by use of a technique known as digital X-ray mapping. In this case a grid is superimposed on the image of the SEM as for the acquisition of digital images, but the signal intensity that is measured and displayed for each pixel is X-ray signal strength for a particular element.

Figure 3 gives an example of an energy dispersive X-ray spectrum. Energy level is plotted on the horizontal axis and the number of detected X-ray events is plotted vertically. The plot shows the general shape of the background (Bremstrahlung) radiation with peaks for sodium, silicon and calcium.

AUTOMATED PARTICLE ANALYSIS INTRODUCTION

The emphasis is on subdivision of features into different «type» particles so as to develop a classification scheme, on the search for exotic inclusions and on the comparison between different samples.

The main purpose of the analysis is to produce a series of descriptions of the types of particle present and a tally of the total numbers and sizes of each type. The amount of information required is defined by the nature of the problem and the innate similarity or dissimilarity between the features. The amount of overlap acceptable is decided for each situation.

The best system is the one that acquires just enough information to perform a separation without redundancy. Where gross differences between materials exist, few parameters are needed. Thus if only one set of particles contains iron, it is only necessary to define one parameter to perform a separation. If the similarity between features increases more variables are introduced.

One method of expressing this is to refer to «like» particles clustering together when plots of different measured parameters are made. «Unlike» particles (i.e. ones which are very different in terms of the plotted variable) will occur in a different area. The human brain can easily interpret clusters in two and three dimensional space – multivariate statistical techniques, including cluster analysis, can perform similar calculations in what can be termed «multidimensional space». The number of parameters measured defines the number of axes of the multidimensional space.

The technique has two main stages – data acquisition and data interpretation. The former includes the establishment of the operating parameters of the SEM (suitably thresholded image to include the required features, appropriate resolution and X-ray acquisition time), the computer

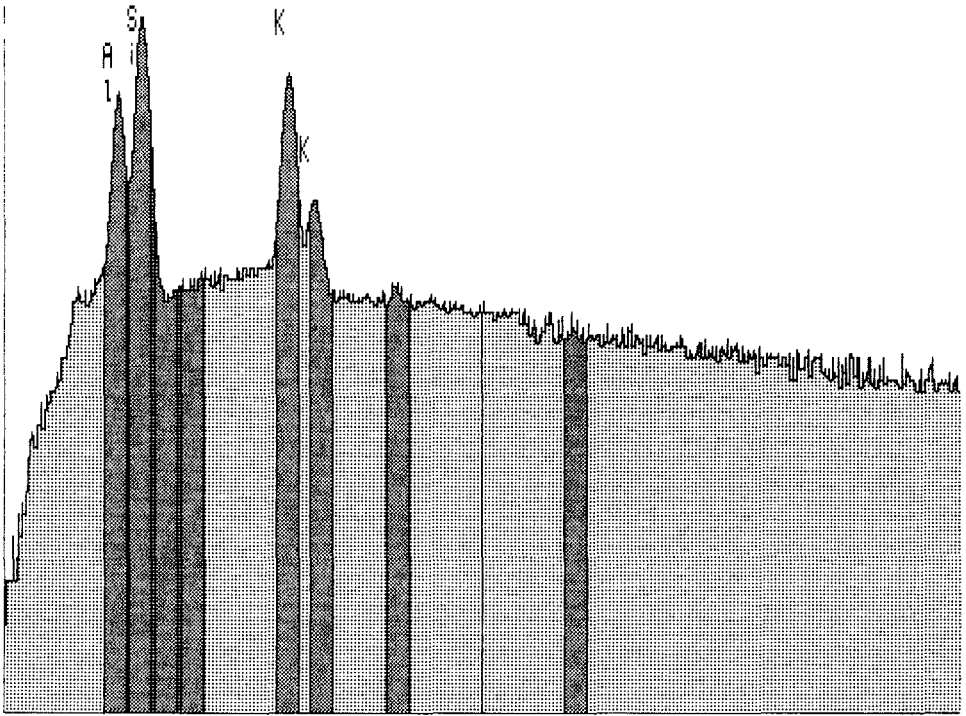


Fig. 3. Example of an energy dispersive X-ray spectrum.

controlled search for features and the measurement of their size and X-ray data. The latter involves statistical and graphical interpretation of the stored data to assess its quality, the establishment of type particles leading to development of a classification scheme, and the use of the results to compare samples. Both are dealt with in separate sections, although they are strongly linked.

PARTICLE ANALYSIS DATA ACQUISITION

A manual examination is normally undertaken as a preliminary phase before automated analysis to allow the operator to familiarize himself with a new sample, to provide a list of the elements present, and to examine the particle distribution on the prepared specimen.

As a summary procedure feature classification requires the description of the various particle types of interest. Objective criteria must be established for the values of each chemical and physical observation parameter. In a manual characterisation mode, the observation variables are portrayed as analogue signals – images, micrographs and X-ray spectra.

The recent development of image analysis software has made it possible to « teach » computers to perform some parts of the analysis. The major advantages of this are that it allows the acquisition of data for large numbers of features in short periods and that the analysis is less dependent on the skill of the operator. It may well also be much more objective. The facility for the creation of large data sets means that a variety of powerful multivariate statistical techniques can be employed to examine patterns and trends that may not be apparent in small, potentially biased samples. One limitation of the automated procedure, however, is that even the most sophisticated software cannot capture the full range of information integrated by the human eye. Therefore the digitized results present a necessarily simplified « picture » ; hence there is a loss of potentially useful information.

Computer control of data acquisition is achieved by use of a digital grey level image as described above. The controlling image analysis program (DIGISCAN) uses this information to locate and size any features present which fall between given (operator selected) threshold values. This image is analogous to a binary switching device with white (« on ») and black (« off »), allowing the computer to count only those pixels (picture points on the image) which are white (on). It scans the image and notes the location of each of these pixels. All touching pixels in a feature are summed to give its two dimensional area. The beam is then driven back under computer control to each feature in turn for the collection of shape and X-ray information. The operator calibrates the system to optimise it for the required balance of speed (throughput) and X-ray data quality by selection of suitable count times and operating voltages and currents.

DATA PROCESSING X-RAY CORRECTIONS

As described above, the energy dispersive X-ray spectrum is represented as a histogram of the total X-ray photons counted by each channel of the multi-channel analyser in the detection system. Each element detectable by the system has one or more characteristic energy levels at which X-rays will be detected if it is present in the sample. A background (Bremstrahlung) radiation distribution is generated by random noise and the elements present show as peaks superimposed on this. These peaks span several channels in the multichannel analyser and a convenient way to summarise the data is to delimit a « region of interest » (often termed « to paint a window » — several are shown as dark stripes on Fig. 3) over the channels concerned and to record the total counts in the whole region. This leads to a significant reduction in required storage space since the counts for each element are now represented by a single number, and it is not necessary to have a number for each channel.

For the automated particle analysis using the DIGISCAN program, up to 25 regions of interest are defined on the spectra and the numbers of X-ray counts falling in each region (or «window») are stored. Several regions are defined on parts of the spectra where no elemental peaks are expected to occur. These are used to subtract the background counts from under each of the other regions of interest which are defined at the energy levels characteristic of the elements which it is desired to measure. The background channel selected and the proportion subtracted form the «background correction».

In certain cases a peak for one element will overlap a peak for another. The «M» line for lead for example, falls in the same region as the «K» line for sulphur. Lead has other lines (the «L series») visible in the 25Kv spectrum, however. Therefore lead is estimated from one of these regions and the number of counts that should occur in the «M» region is predicted and subtracted from the joint peak. The remainder can be designated as sulphur. The proportion of one peak subtracted from another in this manner is termed the «overlap correction». A final correction factor («efficiency factor») allows a linear scaling of the elemental analysis to account for differences in detector efficiency between elements. The set of definitions of X-ray regions of interest and the correction factors to be applied to them, is stored in a separate computer file termed a «window file», since it will be used repeatedly to correct the results for all measured features. Results from a number of analyses must have been obtained using the same window file if they are to be fully comparable.

In this type of semi-quantitative analysis average factors are derived which work reasonably well for the majority of situations. They therefore do not work optimally for every feature and overcorrection may occur leading in some cases to apparent negative concentrations for some elements. These can be set to zero but are commonly retained to allow assessment of the overall data quality. A parameter known as «fit» has been derived to assess this for an individual feature. «Fit» is a scaled ratio between positive and negative values and the larger it is the worse the correction factors are operating. A subset of the data with acceptable values can be chosen. In other cases the apparent negative values contain information that is useful – this relates to the detection of elements which are not specifically included in a window file.

The ability to define 25 regions of interest means that in complex situations choices may have to be made about which elements to include in the window file.

Different size particles of the same material will yield different absolute totals of X-ray counts in each of the specified regions of interest and so the results are normalised by expressing them as a percentage of the total

sum of the counts in all the regions of interest (after correction). This normalisation procedure means that apparent values for an element may appear very similar between two features, where in fact the true percentage is very different. Thus carbon is not detectable but forms a large percentage of the chemical composition of, for example, carbonaceous fly ash spheres. The remaining elements may occur as traces but appear to be major constituents once the normalisation has occurred. Thus a fully quantitative analysis by an appropriate method might give two analyses :

1. Carbon 97 %, Si 1 %, Al 2 %
2. Si 33 %, Al 25 %, K 20 %, Ca 22 % but a normalised analysis of the elements detectable in the current equipment would give an identical Si percentage (33 %). In this type of analysis it is often possible to find other elements on which to distinguish between different particle types.

While it is important to be aware of the uncertainties in the data it should be remembered that the object of the classification procedure is to separate different features from each other and this may often be achieved in spite of the fact that the correction factors have given a distorted apparent composition.

Figure 4 is a « quantile » plot for Si. Such plots are similar to a cumulative frequency plot, except that every point is plotted rather than the central point of grouped data and that the x-axis is divided into standard deviation units rather than percentages. In order to produce the plot the data is first ranked into ascending order. The median value (50 % point on the list) is assigned a quantile score of 0.0, equivalent to the mean, and other values are calculated and plotted.

If the data is normally distributed a straight line plot will be obtained, non-normal data will give curved plots. Most usefully in this context, mixed populations (polymodal distributions) will give inflected curves. Thus it is easy to see that a number of populations are present in this sample.

Figure 5 is a scatter plot for aluminium and silicon. The value for each element in every feature in the sample is plotted. Some obvious « clusters » are apparent and use may be made of these distributions to sub-divide a sample into its constituent parts. Where particles of different types have similar Si or Al values they will overlap on this plot, but may be separable if a different element combination is plotted.

DATA INTERPRETATION CLASSIFICATION

A classification scheme, in this case, is simply a set of particle descriptions (descriptions of those features which define a particle type). When a feature is analysed the computer searches through the list until a

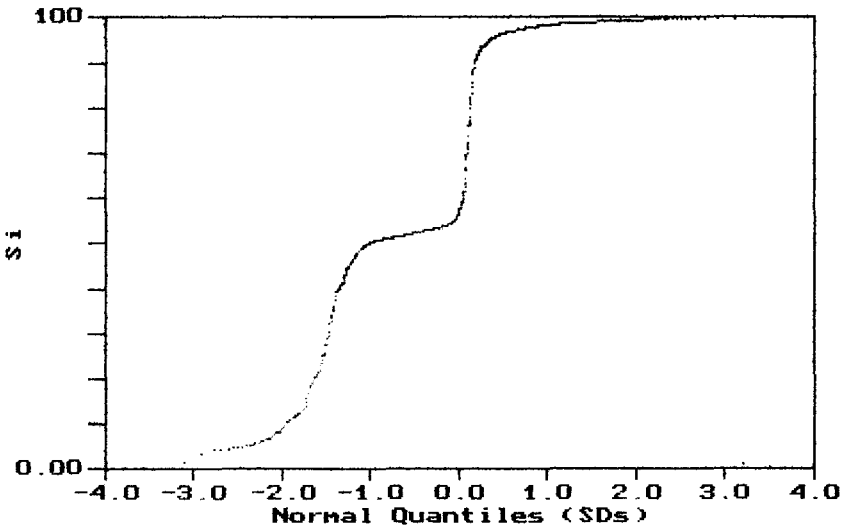


Fig. 4. A « quantile plot » for Si.

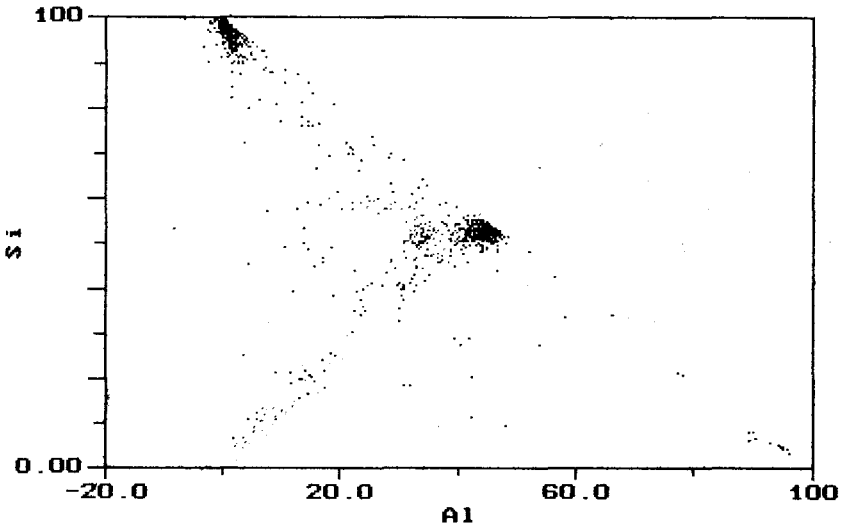


Fig. 5. A scatter plot for aluminium and silicon.

description is found whose conditions are fulfilled. In the present case the feature is then ascribed to that class without further search.

The classes can be developed in several ways. Analysis of a complex unknown sample may be undertaken and the whole data set examined for any obvious groups or associations within it. This is undertaken as follows.

Preliminary subdivisions are made by examination of the distribution of single elements using quantile plots and pairs of elements using scatter plots. Figs 4 and 5 are examples of such plots. In each case the value obtained for the amount of the element in every individual particle in a data set, or combination of data sets, is plotted. Use is made of any obvious discontinuities to split the data into the various populations revealed. The mechanism for subdivision in the scatter plots is to draw boundaries around any obvious clusters and to use these ranges to select a subset of the data. The quantile plots show breaks in slope if subgroups exist in the data set, which can then be split at the point of change. These derived subsets are then examined for all the other stored parameters to produce a description of the particle type (see Johnson *et al.*, 1991 and Sinclair, 1979 for full description of the method).

Subsets of the data can be examined separately to assess the homogeneity of particle types described from plots of the entire data set. If no further potential for subdivision is apparent, the particle type description is added to the classification scheme. This temporary selection feature is particularly useful for examining unclassified particles in isolation as further work may be necessary on them.

Other methods of producing classes include starting with the potential sources and seeking to build up a library of chemical classes each containing the «fingerprint» of a different material which has been examined in isolation — clay mineral standards for example. This also forms a classification scheme and if «unknown» particle types remain then new source type materials have to be examined. Empirical trials of likely element combinations derived from the operator's experience can also be useful.

The plots may show that there are a large number of unclassified features and perhaps highlight the intermediate position of many of these features between clusters of groups that have been classified. Further work would have to be undertaken to assess whether this was due to aggregation of particles of two or more of the described types, or to the presence of as yet undescribed particle types. Quite clearly however this capability of examining distributions and placing unclassified features in a variety of contexts on plots of different element combinations has great power in the assessment of the sample in relation to a classification. Once a scheme has been fully developed it is often the unclassified features that can provide extra information.

DISCUSSION

Scanning electron microscopy is a specialised, capital intensive technique. A method has been introduced which, by greatly improving the rate and duration of analysis, is capable of significantly increasing the number of

particles that can be examined in a given period. Therefore statistical reliability is enhanced. It has also been shown, however, that significant loss of information compared to expert manual analysis, inevitable in these systems. Clearly the technique must be viewed in an appropriate context, and it will not be suitable for all applications. It falls between established bulk analytical methods and traditional scanning electron microscopy and microprobe analysis. The former techniques have evolved to the point where they are rapid, reliable and cheap methods for pinpointing the existence of an anomaly (a mineral deposit, pollution event or heavy metal halo, for example) and for delimiting its severity and extent.

Once a situation is recognized and mapped, however, it is often desirable to discover more detail. In different situations this may involve study of the origins of the phenomenon, the sources from which a deposit has been emitted or evidence towards the provenance of an artefact. In other situations the study may concern process — weathering or other alteration, transport, sorting, deposition or resuspension. Some features may be used as tracers of a particular process — as in the use of fly ash from lake sediments to give evidence for fossil fuel burning as the source of acidification shown by accompanying paleoenvironment reconstruction (cf Rose, this volume). The current influence of particulate material in the corrosion and weathering of buildings, sculpture and other important objects is also clearly of considerable concern to anybody interested in the preservation of our cultural heritage.

This paper has been constructed to describe the capabilities of the technique of individual particle analysis by computer controlled electron microscopy. It has not included any specific examples for reasons of brevity but a number of references are appended to provide actual applications — mainly in environmental studies. Any application which involves particulate material, and especially where this consists of a number of different components, may benefit from consideration of the technique. In addition the SEM has other capabilities not considered here, for example measurement of microtopography, wavelength dispersive X-ray analysis for trace element measurement, mapping of element distributions across a sample, stereoscopic photography — which make it an increasingly important instrument in the fields of archaeology and conservation.

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