Quantitative characterization of individual particle surfaces by fractal analysis of scanning electron microscope images

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Abstract. Morphological characterization of individual particle surfaces was explored by off-line image processing of data obtained by scanning electron microscope – microanalyzer. The fractal geometry was studied by two methods, the power spectrum and the variogram approach. Both methods were evaluated, theoretically by a series of numerically simulated surface profiles and experimentally on a set of pre-recorded secondary electron images of particle surfaces exposing characteristic textures. It was shown that the fractal approach could stand as a base of the methods enlarging the application of electron probe X-ray microanalyzers for individual particle characterization.

Introduction

Electron probe X-ray microanalysis (EPXMA) has for long been extensively used for quantitative measurements of the elemental composition of individual particles. Scanning electron microscopy (SEM) has been used for their visual morphological examination. Quantitative interpretation of surface textures has also been applied for various purpose (see, e.g. [1]). However several facets of texture ought to be mastered quantitatively, a discipline in which so far little progress has been made [2]. Texture quantification would not only lead to reliable surface characterization, it would also extend the knowledge on relating structure with surface properties (i.a. adsorption) and with shaping processes (i.a. physical and chemical weathering). In this respect, direct measurements are becoming increasingly important through the availability of sophisticated image analysis systems and associated software. The recent introduction of fractal analysis as a refinement on the conventional roughness parameters provides a powerful tool capable of revealing systematic differences in texture.

The surface characteristic of particles are important since they are really their ‘fingerprints’. Indeed, the particle surface texture itself results from a combination of alternating chemical and physical processes, leaving markings that reflect more or less the origin of the particles. So far, SEM exoscopic studies [3] allowed, to a certain extent, the reconstruction of the history of individual suspended particles based on their surface texture. An example can be found in the study of suspended quartz grains in the Loire river [4]. This method is however very time consuming which makes the need of an automated direct method through a quantitative description of texture even more necessary. Therefore an attempt was made to extract information on surface morphology of individual microscopic particles by mathematical processing of SEM images. The interpretation of SEM images is clearly of paramount importance if conclusions are to be drawn about the morphological nature of individual particles. A quantitative description of surface texture, based on digital images of surfaces, could eventually contribute to the classification of particles of different morphology, origin or history. In the first instance, this can be done by incorporating the surface descriptors in a form suitable for computer processing off-line. In a later stage on-line processing may become possible.

Up till now the problem of texture quantification essentially lay in the fact that it is very difficult to grasp a concept such as texture and evaluate it by a single parameter, as we intuitively view texture as a measure of several properties such as smoothness, coarseness, heterogeneity and regularity. However, the introduction of a fractal geometry provided a tool to tackle the problem of describing the concept texture. Theoretically a fractal can be defined as a set for which the only consistent illustration of its metric properties requires a dimension value D (called fractal dimension) larger than the standard topological dimension (T):

Dedicated to Professor Dr. Dieter Klockow on the occasion of his 60th birthday

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D = T + (1 - H)

where the parameter H is commonly referred to as the codimension. This practically means that curves can have their ruggedness described by allocating a fractal number between one and two establishing the space filling ability of the curves. Similarly, the concept of fractal dimension can be extended to higher dimensions, e.g. a rugged surface can be given a number between 2 and 3. Assigning fractal dimension values to rugged surfaces therefore allows the texture to be mastered quantitatively rather than qualitatively. Several methods may be applied to obtain the codimension H and thus the fractal dimension D.

In a first method the fractal dimension of the function z(x) may be defined in terms of the Fourier power spectrum P(ω) of the function, as the Fourier power spectrum falls off with increasing frequency ω [5], proportional to:

\[ P(ω) \approx ω^{-(2H+1)} \]

By using a linear regression on the log-log plot of the observed power spectrum (PWS) as a function of frequency, the codimension H and the fractal dimension D can be determined. This method of surface analysis in terms of the power spectrum appears to be so far little explored in the microscopic domain [6] in comparison with the Fourier approach of macroscopic surfaces such as landscapes and environmental topographies [7].

Another method for fractal dimension calculation that was implemented is the so-called variogram method. This method can be defined in terms of how the variance of interpixel differences changes with distance. The fractal dimension of the function z(x) may then be estimated from a log-log plot of the variance of increments versus increments:

\[ \text{Var}(x) \approx x^{2H} \]

The resulting graph is called a variogram, from which the name variogram method of this fractal dimension calculation procedure originates. The variogram method has been extensively adapted to analyze a number of environmental data (from anemones to rainfall). A review of the applications has been given by Burrough [8].

Other experimental methods for fractal-based characterization of surface texture exist, but are more complicated being mainly based on the different sectional approaches and are beyond the intention of this paper. The vertical section method comprehends for example the structured walk technique [9] or the box dimension approach [10] on vertical sections to the fracture plane, whereas the slit island technique [6] makes use of sequentially prepared sections parallel to the fracture plane and is based on the perimeter-area relation.

**Experimental**

**Image acquisition**

The images are recorded and stored with the Tracor Northern TN 2000 and TN 1310 automating system of a JEOL 733 electron microprobe. The images collected for our purposes were 256 analysis pixels in both x and y directions. All images were collected in the secondary electron mode because the secondary electron images generally show very low noise level and a sharp contrast with high spatial resolution [11]. The absolute signal intensity at each analysis pixel was converted from an analogue to a digital signal by an 8 bit resolution analogue to a digital converter (ADC). The system is thus capable of coding the images in as much as 256 brightness levels. Since the system does not offer adequate bulk storage and processing facilities, the images are stored on floppy disk or on tape and transferred to VAX/VMS minicomputer for further processing.

**Image processing**

To constraint the processing time and to simplify the interpretation for the power spectrum method, the one-dimensional version of the fast Fourier transform (FFT) was preferentially applied. In this one-dimensional approach, the FFT was run consecutively on the 256 transect lines composing the image. The PWS was calculated by recombining the imaginary and real part of the transform to establish real data. Subsequently the 256 power spectra, taken from serial sections, were averaged to reduce the influence of microstructure and statistical artefacts. This average PWS was finally transformed into a log-log plot before exercising a linear regression fit routine. The fractal dimension was estimated from the slope, considering the following equation:

\[ P(ω) \approx ω^{-(5 - 2D)} \]

The variogram method of measuring the fractal dimension has the advantage that it is an easy method in application, which does not involve too difficult mathematics and which does not require any preceding data adaptation. Additionally, it was based on good statistics since the method was applied in its two-dimensional version, i.e. on the whole data matrix simultaneously. Hence the occurrence of possible problems due to directionality of the surface is also excluded. Since it is more convenient, the standard deviation was calculated and used instead of the variance. Nevertheless, the term variogram will be maintained because it is commonly used in the literature. For this method, it was sufficient to scan the whole 256 × 256 matrix, to calculate the differences in image intensity, in the horizontal and the vertical direction. Starting with a pixel step size equal to 1, the procedure was repeated for successively increasing pixel step increments (up to 200). The distributions of the intensity differences were approximately Gaussian, with zero mean, for all increment settings. The standard deviation σ(k) value corresponding with a certain pixel step k was calculated by recombining the individual sums of squared differences respectively for the x and y directions. The whole procedure was repeated for a limited number of pixel distances up to 200, because for higher pixel distances the statistics readily deteriorate. Also at higher pixel steps, only differences in the boundaries will be measured. Subsequently the standard deviation was plotted as a function of the pixel step in a log-log graph. From the linear least-