# Chapter 3 SCANNING ELECTRON MICROSCOPY, IMAGE ANALYZER, AND STEREOLOGY

## 3.1 INTRODUCTION

The microscope has been a powerful tool in the study of cement and concrete since the early development of these materials. Le Chattelier (1882) was amongst the first to apply the microscope to the study of cementitious materials. He used it to investigate the chemical and physical aspects of hydration and setting, rather than to study cracks. His efforts undoubtedly influenced later workers in their use of the microscope. Tavasci (1942) successfully used the microscope to study the composition and structure of concrete, but not for cracks per se. His work, however, set the stage for the studies of cracks on the interior surfaces of cut specimens which were conducted in the 1960s.

The electron microscope was apparently first used by Eitel (1941, 1942), and by Radczewski and his co-workers (1939) to study the hydration process of concrete. Grudemo (1960) was another important pioneer in the use of high magnification, including the use of the electron microscope. Although most of these studies were not directly related to cracks, they led the way to later studies of cracks in which electron microscopy was a powerful tool. Diamond and Mindess (1980) used the scanning electron microscope to observe the growth of surface cracks during loading, using magnifications generally from 35X to 450X.

### 3.2 SCANNING ELECTRON MICROSCOPY

The scanning electron microscope (SEM) is one of the most versatile instruments available for the examination and analysis of the microstructural characteristics of solid objects. The primary reason for the SEM's usefulness is the high resolution that can be obtained when bulk objects are examined; values on the order of 2 to 5 nm (20-50 Å) are now usually quoted for commercial

instruments, while advanced research instruments are available that have achieved resolutions of better than 1 nm (10Å).

The basic components of the SEM are the lens system, electron gun, electron collector, visual and recording cathode ray tubes (CRTs), and the electronics associated with them. In the SEM, a fine electron probe is produced which rapidly scans (rasters) across the area of interest. The signals generated in the latter case are detected and converted to CRT electronic signals, which are then fed to a cathode ray tube (CRT). The CRT and the scanning coils are linked through the same scan generator, so that the image appearing on the CRT corresponds spatially to the area of the sample scanned.

The interaction of the electron beam with the specimen produces a variety of signals that are used for imaging and spectroscopy. These signals are not generated at a point, but rather within a volume known as the *interaction volume*.

The incident (primary) electrons lose energy as they penetrate the sample, giving rise to an X-ray continuum, which consists of all possible wavelengths corresponding to the range of energies of the incident beam. The high-energy primary electrons may penetrate some distance into the sample before being scattered outside of the sample again by the Coulombic repulsion of the electron clouds in the solid. It has been experimentally determined that a significant fraction of the incident electrons that strike a flat, bulk target placed normal to the probe subsequently escape through the same surface that they entered. The re-emergent beam electrons are called backscattered electrons (BSE). The strength of the scattering will depend on the atomic number of the scattering atom, so that backscattered electron images exhibit atomic number contrast. Backscattered electrons provide an extremely useful signal for imaging in scanning electron microscopy. Backscattered electrons respond to composition (atomic number or compositional contrast), crystallography (electron channeling), and internal magnitude fields (magnetic contrast). Note that these backscattered electrons may be generated at greater depths than that indicated, in which case they may not possess enough energy to escape the sample. The primary or backscattered electrons may knock electrons out of the conduction band of the solid. These secondary electrons are relatively low in energy, and so can only escape from a

region near the surface of the sample. This signal is thus often used for generating topographic information.

The electron probe and the CRT are linked through the same scan generator, so that both sample and screen are scanned in the same X-Y grid pattern. The intensity of the signal reaching the detector from a given point on the sample is used to adjust the brightness of the CRT at the corresponding point. The result is the construction of a map of the sample. Figure 3.1 shows a scanning electron microscope and an image analyzer.





Figure 3.1 Scanning electron microscope (SEM) and Kontron Image Analyzer

# 3.3 IMAGE ANALYZER

The backscatter electron images obtained from scanning electron microscopy were analyzed on *Kontron Electronik GmbH Image Analysis Division, IBAS* "*Interaktives Bilt-Analysen System*" (*Interactive Image Analysis System*). Computer programs were developed to analyze the images based on the concept of stereology which is covered in detail later on in this chapter, and also based on two dimensional measurements of the crack network represented by Wood's metal. The computer program for stereological analysis obtains fifteen parameters from each image. The area of the image that Kontron analyzes is a square with the dimensions  $512 \times 512$  pixels, each pixel corresponding to 3.2890 microns. The program creates a histogram for each image and establishes a threshold in the histogram, in order to identify the Wood's metal. It then scraps the noncontinuous short objects, lines, and dots out of the image. Its next step is to thin the features in the scraped image to create a binary image. The binary image is then intersected by an array of straight parallel lines at various angles, in this case at angles of 0°, 15°, 30°, 45°, 60°, 75°, 90°, 105°, 120°, 135°, 150°, and 165°. The number of crack intercepts at a given angle is measured according to the number of intersections on line array at that angle with the features in the binary image.

# Number of Crack Intercepts( $\theta$ ) = FIELDCOUNT

The program also calculates the total area of the features in the binary image. Since the area of each image is known, the percent of the area that is cracked (or, the crack density) can be determined. For each parameter in the computer program, two values were calculated: the first relates to the final binary image from the original image, and the second relates to the smoothed version of the cracks. Figure 3.2 is a flow chart summarizing the computer program for stereological analysis. The computer programs are presented in Appendix 2.



**Figure 3.2** Flow chart for stereological steps

# **3.4** STEREOLOGY AND CONCRETE

#### 3.4.1 INTRODUCTION

All matter can be described in terms of zero, one, two, and three dimensions. Stereology deals with the interpretation of three-dimensional structures by means of their two-dimensional sections. Stereology is the opposite of photogrammetry, which utilizes three-dimensional images in order to construct flat maps. Techniques conventionally used for studying the three-dimensional structure of materials, particularly in other material sciences, are often stereological ones.

If a sectioning plane cuts a three-dimensional aggregate of space-filling polyhedrons, a two-dimensional structure that consists of area-filling polygons can be observed. The task then is to relate the observations made on a section to the true three-dimensional microstructure. Stereology attempts to characterize numerically the geometrical aspects of those features of the microstructure of interest; for example, the microcracks in concrete represented by Wood's metal. In its broadest context, stereology includes not only the quantitative study and characterization of any spatial structure, but also its qualitative interpretation.

There are various approaches to stereological problems. The statisticogeometrical approach depends on measuring and classifying a large number of two-dimensional images and is the method utilized in this study. It is applicable when objects are randomly distributed in space. In such cases, a single section, if extensive enough to contain a statistically significant number of features, may suffice to obtain valid results.

In this study, we deal with the numerical or quantitative characterization of points, lines, surfaces, and volumes. Fundamental expressions have been determined which relate measurements on two-dimensional sections to the three-dimensional structure.

## **3.4.2 BASIC MEASUREMENTS**

Table 3.1 presents some of the basic symbols commonly used in the measurements employing quantitative stereology.

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Symbol	Dimensions	Definition
Р		Number of point elements, or test points
$P_L$	μm <sup>-1</sup>	Number of intersections of cracks in a section with a superimposed system of equally spaced test array of straight parallel lines per unit of line length
$P_L( heta)$	μm <sup>-1</sup>	Number of intersection of cracks in a section with a system of equally spaced test array of straight parallel lines positioned in such a way that it successively encloses an angle $\theta$ , $\theta = \pi/2$ and $\theta = 0$ , respectively, with the axis of symmetry
L	μm	Length of lineal elements, or test line length
$L_A$	μm/μm²	Total crack length in a section per unit of area
Α	μm <sup>2</sup>	Planar area of intercepted features, or test area
A <sub>A</sub>	μm²/μm²	Area fraction. Area of intercepted features per unit test area
S	μm <sup>2</sup>	Surface or interface area (not necessarily planar)
$S_{V}$	μm²/μm³	Total crack surface area per unit of volume $\left(\frac{S}{V_T}\right)$
N <sub>A</sub>	μm-2	Number of cracks in a section per unit of area
V <sub>T</sub>	μm <sup>3</sup>	Volume of three-dimensional features, or test volume

Table 3.1 List of basic stereological symbols and their definition

In this research, the stereological parameters of  $P_L$ ,  $L_A$ , and  $S_V$  will be used to perform the stereological analysis. The derivation of the relationships between  $L_A$  and  $S_V$  with  $P_L$  is presented below (Underwood 1968).

# 3.4.2.1 Number of Point Intersections, P<sub>L</sub>

A linear test array is applied randomly to the microstructure in the section plane.  $P_L$  is the number of points (intersections) generated per unit length of test lines.

#### 3.4.2.2 Surface-to-Volume Ratio, S<sub>V</sub>

The aim of this method is to obtain the surface-to-volume ratio of a system of surfaces in a volume. The basic equation for obtaining the area of surfaces in a volume is

$$S_V = 2P_L \ \mu m^2 / \mu m^3$$
 (3.1)

which was derived by Salitikov (1945) and later by Smith and Guttman (1953).

Equation 3.1 applies to a system of surfaces with any configuration. It is as valid for systems of interconnected surfaces as for systems of discontinuous, separated, or bounded surfaces.

In order to derive Equation 3.1, let us consider a test cube of edge length l and volume  $V_T = l^3$  enclosing a system of surfaces oriented randomly throughout the cube. The surfaces may be planar or curved, continuous or interrupted, isolated or connected, as represented in Figure 3.3. A set of N vertical test lines of total length  $L_T = Nl$  is passed randomly through the cube, cutting horizontal planes through the cube with density  $N/l^3$ .



**Figure 3.3** Model of deriving the relationship  $S_V = 2P_L$ . This cube contains random surfaces cut by random vertical test lines (Underwood 1968)

The total surface *S* is divided into *n* elementary units of surface area  $\delta S$  so that  $S = n\delta S$ . The normals to each elementary area form angle  $\theta_i$  to the vertical test lines, and the areas of the projections of the elementary areas on a horizontal plane, equal  $\delta S \cos \theta_i$ . Thus the fraction of test lines intersecting the elementary areas is  $\delta S \cos \theta_i / l^2$ .

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An expression is set for the density of intersections of the test lines with the elementary areas, then the individual contributions are added to obtain the total number of intersections with the total surface area. If  $P_i$  is the number of intersections associated with each elementary area, then the expected value of the total number of intersections with the entire surface is

$$E(P) = \sum_{i=1}^{n} P_{i} = N \sum_{i=1}^{n} \frac{\delta S \cos \theta_{i}}{l^{2}} = \frac{N \delta S}{l^{2}} \sum_{i=1}^{n} \cos \theta_{i}$$
(3.2)

Since the elementary units of surface are oriented randomly, every value of  $\theta_i$  has equal likelihood and  $\left(\frac{1}{n}\right)\sum_{i=1}^{n}\cos\theta_i$  equals the average value,  $\overline{\cos\theta}$ . Making appropriate substitutions in Equation 3.2 yields:

$$E(P) = \left(\frac{N}{l^2}\right) (\delta Sn) \overline{\cos \theta}$$
(3.3)

Since  $n\delta S = S$ , and  $\frac{N}{l^2} = \frac{Nl}{l^3} = \frac{L_T}{V_T}$ , where  $L_T$  is the total length of test arrays, Equation 3.3 can be rewritten as:

$$E(P) = \left(\frac{L_T}{V_T}\right) S \overline{\cos\theta}$$
(3.4)

where E(P) is the expected value of the total number of intersections of the test lines with the system of surfaces.



**Figure 3.4** Geometry involved in the determination of the probability that random normals lie between  $\theta$  and  $\theta + d\theta$  (Underwood 1968)

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The evaluation of  $\cos \theta$  can be visualized by means of the hemisphere depicted in Figure 3.4. The probability that normals lie between  $\theta$  and  $\theta + d\theta$  is expressed by

$$P(\theta)d\theta = \frac{\text{areaof zone}}{\text{areaof hemisphere}} = \frac{2\pi r^2 \sin\theta d\theta}{2\pi r^2} = \sin\theta d\theta$$

and average value of  $\cos\theta$  is

$$\overline{\cos\theta} = \int_{0}^{\pi/2} \Pr(\theta) \cos\theta d\theta = \int_{0}^{\pi/2} \sin\theta \cos\theta d\theta = \frac{\sin^2\theta}{2} \Big|_{0}^{\pi/2} = \frac{1}{2}$$

Rearrangement of Equation 3.4 gives

$$E(P) = \frac{L_T}{V_T} \frac{S}{2}$$
 or  $\frac{S}{V_T} = \frac{2E(P)}{L_T}$ 

which yields the required relationship, in our notation,

$$S_{\rm V} = 2P_L \ \mu {\rm m}^2 \ / \ \mu {\rm m}^3$$
 (3.5)

### 3.4.2.3 Length of Line Per Unit Area, L<sub>A</sub>

The equation that relates the length of lineal elements in a plane to their intersection with a test line is

$$L_A = \left(\frac{\pi}{2}\right) P_L \quad \mu m / \mu m^2 \tag{3.6}$$

The quantity  $L_A$  is a basic microstructural parameter that is useful either as it is or when manipulated into other forms. The derivation of Equation 3.6 is similar to the derivation of Equation 3.1.

Given a randomly oriented system of lines in a plane, let us consider a square test area  $A_T$  of edge length *l*. A set of *N* vertical test lines of total length  $L_T = Nl$  is passed randomly through the test area, cutting horizontal lines through the square with density N/l. The system of lines is divided into *n* straight elementary segments of length  $\delta L$  and the total line length in the system is

 $L = n\delta L$ . The elementary segments form angle  $\theta_i$  to the vertical test lines, and the length of projections of the elementary segments on a horizontal line, are  $\delta L \sin \theta_i$ . Thus the fraction of test lines intersecting the elementary segments is  $\delta L \sin \theta_i / l$ .



**Figure 3.5** Model of deriving the relationship  $L_A = (\frac{\pi}{2})P_L$ 

If  $P_i$  is the number of intersections of each elementary segment by the test lines, then the expected value of the total number of intersections with the entire system of lines is

$$E(P) = \sum_{i=1}^{n} P_{i} = N \sum_{i=1}^{n} \frac{\partial L \sin \theta_{i}}{l} = \frac{N \partial L}{l} \sum_{i=1}^{n} \sin \theta_{i}$$
(3.7)

Since the elementary systems are oriented randomly, each value of the angle  $\theta_i$  has equal likelihood of existence and  $\left(\frac{1}{n}\right)\sum_{i=1}^{n}\sin\theta_i$  equals the average value  $\overline{\sin\theta}$ . Making appropriate substitutions in Equation 3.7 yields:

$$E(P) = \frac{N\delta Ln}{l}\overline{\sin\theta}$$
(3.8)

Since  $n\delta L = L$  and  $\frac{N}{l} = \frac{Nl}{l^2} = \frac{L_T}{A_T}$ , Equation 3.8 can be rewritten as:

$$E(P) = \frac{L_T}{A_T} L \overline{\sin \theta}$$
(3.9)

The evaluation of  $\overline{\sin \theta}$  can be visualized by means of the circle depicted in Figure 3.6.



**Figure 3.6** Geometry involved in the determination of the probability that elementary segments lie between  $\theta$  and  $\theta + d\theta$ 

The probability that the elementary segments are oriented between  $\theta$  and  $\theta + d\theta$  is equal to the fraction of the perimeter of a circle that is occupied by this orientation range. From the symmetry involved, only one quadrant of the circle needs to be considered, giving for the probability

$$P(\theta)d\theta = \frac{\text{fraction of perimeter}}{\text{perimeter of circle}} = \frac{rd\theta}{\pi r/2} = \frac{2}{\pi}d\theta$$

Thus the average value of  $sin\theta$  is

$$\overline{\sin\theta} = \int_{0}^{\pi/2} \mathbf{P}(\theta) \sin\theta d\theta = \frac{2}{\pi} \int_{0}^{\pi/2} \sin\theta d\theta = \frac{2}{\pi}$$

Rearrangement of Equation 3.9 gives

$$\frac{L}{A_T} = \left(\frac{\pi}{2}\right) \frac{E(P)}{L_T} \quad \mu m/\mu m^2$$

or, in our notation,

$$L_A = \left(\frac{\pi}{2}\right) P_L \quad \mu m / \mu m^2 \tag{3.10}$$

#### 3.4.3 DEGREE OF ORIENTATION

In real structures, the lines in a microsection of a given specimen are usually either isometric or partially oriented. Only in rare cases do we find completely oriented systems of lines. In a partially oriented system of lines in a plane, part of the total length of lines is oriented in a definite direction (or directions). The remaining segments may essentially have a random orientation.

The lines of a partially oriented system of lines in a plane can be divided into elementary straight segments that are very small and of equal length. Some or all of the segments will lie parallel to one or more definite directions (the orientation axes). The remaining segments are assumed to be oriented randomly, or isometrically. From this point of view, a partially oriented system of lines may be regarded as consisting of two superimposed systems of lines: an oriented portion and a random portion.

The length of linear elements in a plane is proportional to the number of intersections made with a test line. Using a test array of straight parallel lines, the number of intersections with an oriented system of lines (such as a crack system) will vary with the direction of the test array. The dependence of the number of intersections per unit length with the angle of the test array can be used to characterize the degree and type of orientation of a system of lines in a plane.

If a test array system of equally spaced, straight parallel lines is superimposed on a sample area, i.e., an image of the crack network in concrete, the number of intersections per unit length of the test line  $P_L$  can be determined. Since the value of  $P_L$  is a function of the direction of the line system, the specific number of intersections is indicated as  $P_L(\theta)$ . The *rule of total projections* (Stroeven 1973) states that this value of  $P_L(\theta)$  equals the value of the total projected length  $L_{A_{Proj.}}$  of the lineal features upon a line perpendicular to the test array, or

$$P_L(\theta) = L_{A_{\text{Pr},ai}} \tag{3.11}$$

This type of sampling procedure is called the *method of directed secants on a plane* (Saltikov 1967).

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In the case of an isometric (randomly oriented) system, the value of  $P_L(\theta)$  is dependent on  $\theta$  and both sides of equation 3.11 can be averaged with respect to orientation. The result is an important relationship connecting the length of lineal features in a sampling area,  $L_A$ , to the specific surface area or to the number of intersections per unit length of the test line, and averaged with respect to the orientation. Therefore,

$$\frac{\pi}{2}P_{L} = L_{A} = \frac{\pi}{4}S_{V}$$
(3.12)

Applying the method of random secants on a plane to an image of a crack pattern, Equation 3.12 presents simple algebraic relationships to calculate the total crack length per unit area or the specific surface area (of the cracks) per unit volume. The dependence of the number of intersections per unit length with the angle of the test array can be used to characterize the degree and types of orientation of a system of lines in a plane. Saltikov (1945) proposes a polar plot of  $P_L$  with respect to the orientation axis (axes), and calls the resulting curve the *rose of the number of intersections*, or simply the *rose*.

The rose for an oriented system of lines can readily be obtained experimentally by applying a test array to the system of lines at equal angular increments with respect to the orientation axis, and determine  $P_L$  separately at each angle. A polar diagram can be made by plotting the radius vectors,  $P_L$ , versus  $\theta$ . The rose diagram is created by connecting the ends of the radius vectors by lines or a smooth curve. In the case of isometry, the rose will be a circle with its center at the origin of the polar figure. If a preference direction should occur in a crack pattern, the shape of the rose will change.

The stress-induced microcrack system in concrete, a composite material, is considered to be a partially oriented as opposed to a completely oriented (idealized) system.

# 3.4.4 APPLICATION OF STEREOLOGY TO CONCRETE FRACTURE MECHANICS

Stroeven (1973, 1976, 1979, 1991, 1992), Ringot (1988), and Massat et al. (1988) successfully applied the concept of stereology to study micromechanical aspects of concrete. With the advent of modern image analysis systems, it is now

possible to perform stereological analysis on a great number of images accurately and expeditiously, whereas in the past this was not achievable by means of manual methods.

In this section, the application of stereology to the microstructure of concrete will be explained sequentially. After the concrete samples were prepared for scanning electron microscope studies (see Chapter 2), 55 images were extracted from each sample. A total of four samples taken from the center and edge of the concrete cylinders in axial direction were studied (samples 1 through 4 in Figure 2.25). SEM produces a multiphase image from each observation (Figure 3.7).



**Figure 3.7** A SEM backscatter image

Figure 3.8 Establishing threshold in histogram

In order to recognize and isolate Wood's metal, which is the representative of pores and fractures in concrete, the image analyzer can make a histogram for all of the different phases in the image based on their gray levels, with zero representing the darkest phase and 255 representing the brightest phase (Figure 3.8).

From this histogram, and by means of the trial and error method, two threshold levels can be established to encompass the brightest phase in the

image, namely Wood's metal. The threshold for Wood's metal identification was set between 170 and 255 (Figure 3.9).



**Figure 3.9** Wood's metal identification by establishing threshold levels in histogram

The next step is to eliminate objects from the background that don't fall between these threshold levels (*dis2lev* command). Once the above task is accomplished, what is left in the image is the crack network and pores shown by Wood's metal (Figure 3.10). At this point the aim is to eliminate objects on the basis of their area in pixel units. The lower and upper limits of the objects to be eliminated has to be established to include small pores, small non-continuous cracks, etc. (Figure 3.11).



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The next step is to transform this image into a skeletonized binary image by means of a binary thinning process (invoked by *thinbin* command). For every thinning step, pixels that are not relevant to the connectivity of an object are removed from the object margins, i.e., converted into background pixels. The connectivity of objects is thus maintained. This process can be continued until all objects are reduced to a width of one pixel that approximates the skeletons. Figure 3.12 is the final binary image used for stereological measurements.



Figure 3.12 Binary-thinned image of the crack network in concrete

The binary image in Figure 3.12 is then intersected by an array of straight parallel lines at 15° angular increments, in this case at angles of 0°, 15°, 30°, 45°, 60°, 75°, 90°, 105°, 120°, 135°, 150°, and 165° (Figure 3.13). The number of crack intercepts at a given angle is measured according to the number of intersections on line array at that angle with the features in the binary image, or crack

network. The number of intersections were determined separately at each angle  $\theta$ . Then the number of intersections were plotted versus  $\theta$ , creating the rose of the number of intersections diagram. The rose diagram characterizes the degree of orientation of the cracks, and makes it easier to interpret the data.

Number of Crack Intercepts( $\theta$ ) = FIELDCOUNT

The rose of the number of intersections diagrams for all the experiments are presented in Chapter 4. It is clear from these diagrams that the cracks are not randomly oriented and that more lie at angles between 0° and 15° and 165° and 180° than in other directions.

The rose diagrams were plotted to cover only the range of 0° to 165° (cracks at 0° and 180° have equal lengths) since the range from 180° to 360° is redundant (Russ 1986). Figure 3.13 depicts the array of straight parallel lines at 0° (or 180°), 15°, and 165°.



Figure 3.13 Array of straight parallel lines