# LARGE SCALE POINT COVARIANCE ANALYSIS OF HOMOGENEITY IN TiB$_2$-Fe COMPOSITES

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## ABSTRACT

Image analysis of a sintered TiB$_2$-Fe hard material has been performed. The porosity dispersion is discussed from size and covariance function measurements. An extension of the usual point covariance analysis was made by implanting series of 10 contiguous segmentation fields, and making measurements of the linear covariance function in the direction of field alignment. The maximum correlation distance of the analysis was then increased by a factor of 10, as compared to a usual analysis with the same resolution. The method is an alternative to the regularized covariance analysis (Hersant and Jeulin, 1976).
A two scale boolean model is proposed to interpret variations of the measured covariance function from the origin to its asymptotic limit. Using the model, porosity can be represented either by 20-30 $\mu$m clusters, or by the complementary phase of 30-50 $\mu$m (grains+binder) clusters, which should have been formed during the elaboration process.

Keywords: boolean model, covariance, dispersion, homogeneity, sintered materials.

## INTRODUCTION AND EXPERIMENTAL PROCEDURE

Analysis of homogeneity of phase repartition in polyphased materials is doubly important :
at first to understand the influence of the elaboration process on the microstructure of the resulting material, then to forecast physical properties of the material.
Characterization of phase dispersion has been performed on a section of a TiB$_2$-Fe sintered body, with 20 vol% Fe binder. Particular attention has been given to the residual ~3 vol% porosity which subsists in the material although O, C and N impurities have been reduced by NdNi$_5$ and TiFe$_2$ additives (Ottavi et al., 1992).The aim of the study was to give an explanation to this porosity which cannot be eliminated, even after optimisation of the elaboration process. Only three "phases" were segmented for commodity : the TiB$_2$ granular phase (G), arbitrarily including the Ti(O,C,N) phase, the binder Fe (+Ni, Ti) phase (B), arbitrarily including the finely dispersed Nd$_2$O$_3$ phase, and the porosity (P).

Preliminary observations of the sample let us presume multiple repartition scales of the porosity, from the ~2 $\mu$m intergranular spacing to several hundred micrometers. Therefore, a usual point covariance analysis could not give a global description of the microstructure. An extension of this method was performed by implanting series of 10 contiguous segmentation fields, and measuring the linear covariance function in the direction of fields alignment. Analysis was made at low magnification (x 300), in order to characterize large scale fluctuations of the composition, and at high magnification (x 900), to ensure a granulometric analysis with sufficient resolution. Figure 1 is a typical view of the material.
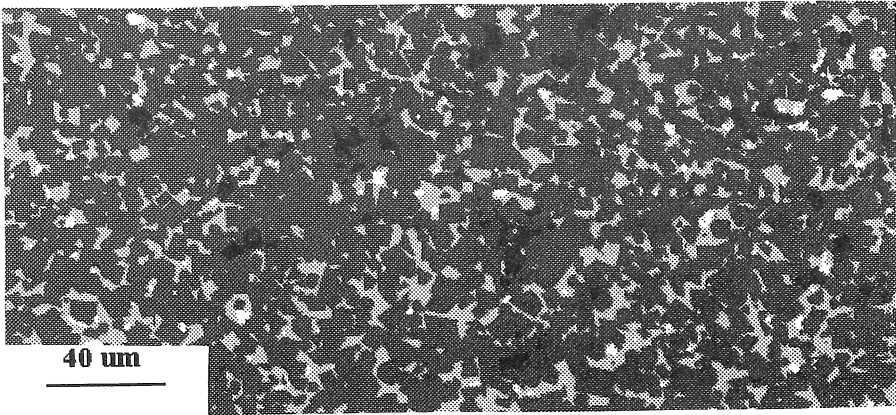
**Figure 1 :** Typical section of a $TiB_2$-Fe material (x 300)


## RESULTS AND MODELING

### 1. CHORD LENGTH DISTRIBUTION

Figure 2 shows that the chord length distributions in the binder phase and in the porosity are roughly linear on a logarithmic scale, with similar slopes, while the one of the granular phase (not shown) is quite different. This indicates that pores tend to replace the binder phase in the microstructure. The exponential distribution shows that the binder phase can be described, in a first approximation, as the complementary phase of a boolean set (Coster and Chermant, 1985), representing the granular phase. With this assumption, the mean chord length $L_1$ in the binder phase and in the porosity can be estimated from the slope of the curves in logarithmic scale (Table 1). The mean chord length in the granular phase is then deduced from the relation :

$$\frac{L_1(G)}{L_1(B + P)} = \frac{V_V(G)}{V_V(B + P)} , \qquad (1)$$

where $L_1(B + P)$ is estimated by averaging on the two phases. The method allows to estimate precisely $L_1$ on images with poor resolution, which is the case of our analysis at low magnification, where large fields are necessary. The values obtained are lower than the direct evaluations, and fit better those obtained at higher magnification (Table 1).
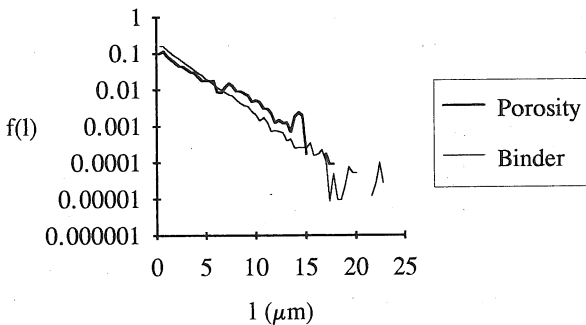


**Figure 2 :** Chord length distribution in the porosity and binder phase.

**Table 1 :** Measurements of volume fractions and mean chord lengths.

|  | Grains | Binder | Porosity |
|---|---|---|---|
| $V_V$ | 0.74 | 0.23 | 0.03 |
| $L_1$ (x 300 : measurement) | 8.4 $\mu$m | 2.7 $\mu$m | 4.3 $\mu$m |
| $L_1$ (x 300 : extrapolation) | 6.7 $\mu$m | 2.3 $\mu$m | 2.9 $\mu$m |
| $L_1$ (x 900 : measurement) | 6.4 $\mu$m | 2.2 $\mu$m | 2.9 $\mu$m |

## 2. COVARIANCE ANALYSIS

Large scale covariance analysis was performed on two series at low magnification (x 300). It shows fluctuations of the different compositions with a period of several hundred micrometers (Fig 3). These fluctuations are strongly dependent on the position of the image series, as can be shown by the comparison of the two series. Their complete characterization (size, amplitude) would need an averaging on a great number of series.
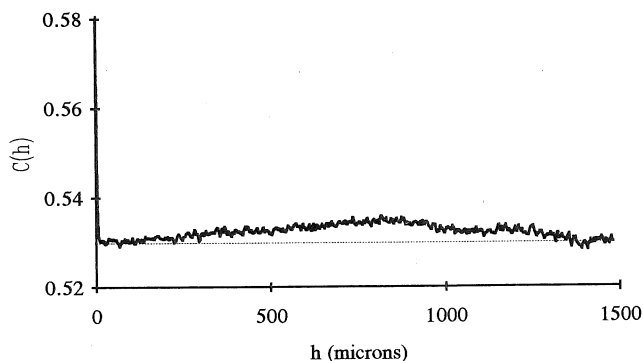


**Figure 3 :** Covariance function of the grains at large scale (x 300).

At small scale, the covariance functions of the granular and binder phases regularly decrease to their asymptotic limit, while the one of porosity exhibits an intermediate stage in the range 10-50 $\mu$m (Fig 4).
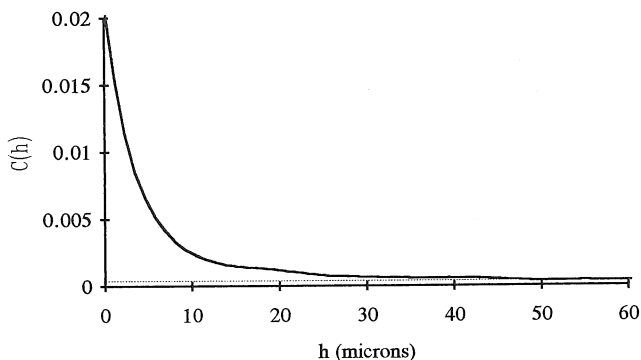


**Figure 4 :** Covariance function of the porosity at small scale (x 300)

Hence, though porosity tends to replace the binder phase, its repartition shows a correlation scale which is greater. This is also evidenced by the integral range $a_1$, which is a mean correlation scale (Jeulin, 1979), and which is higher for the porosity than for the binder phase (Table 2).

**Table 2 :** Integral ranges $a_1$ deduced from covariance function measurements (x 300).

|  | Grains | Binder | Porosity |
|---|---|---|---|
| $a_1$ | 4.3 $\mu$m | 4.0 $\mu$m | 8.3 $\mu$m |

The integral range is a global parameter, which averages an elementary scale of correlation of the porosity (about the same as for the binder or grains), and a greater one, which is evidenced by the intermediate stage at 10-50 $\mu$m. In order to separate these two scales, a description of the microstructure is proposed which uses the intersection of two boolean sets. The method is similar to the one used by Jeulin (1979).
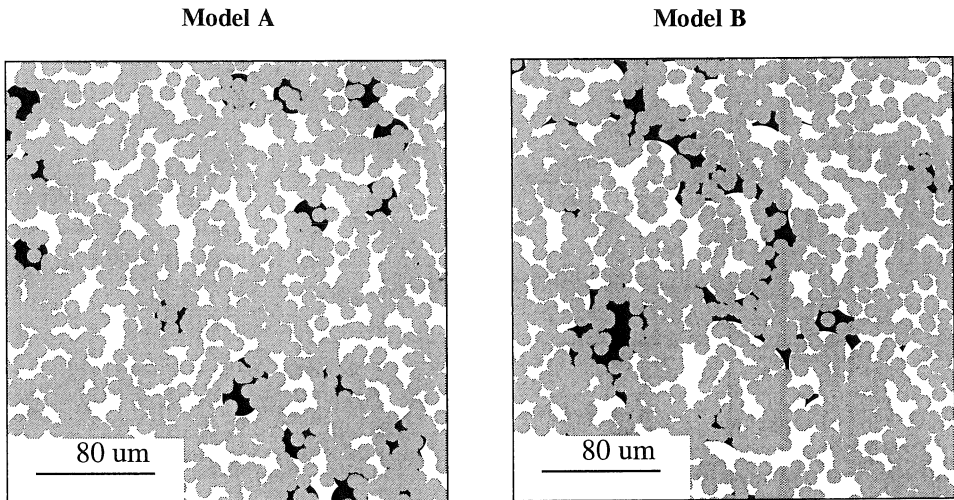
**Model of microstructure**

Model A                                                     Model B



**Figure 5 : 2D representation of model microstructures of porosity in TiB$_2$-Fe material**

A boolean set X is constructed by implanting primary grains of known size and shape at points distributed at random in space. If the volume fraction of grains is $V_V$, the covariance function of such a set and of its complementary are given by (Jeulin, 1979) :

$$C_X^c (h) = \exp \left( \text{Ln} (1-V_V) \frac{2 K(0)-K(h)}{K(0)} \right) , \qquad (2)$$

$$C_X (h) = 2 V_V - 1 + C_X^c (h) , \qquad (3)$$

where K(h) is the geometric covariogram of a primary grain. For a sphere of size D

$$K(h) = \frac{\Pi D^3}{6} (1 - \frac{3}{2} \frac{h}{D} + \frac{1}{2} \frac{h^3}{D^3}) . \qquad (4)$$

Let us now consider a boolean set $X_1$ with volume fraction $V_V(X_1)$, whose primary grains are spheres representing the $TiB_2$ grains, and a second boolean set $X_2$ whose volume fraction is $V_V(X_2)$ and size of spheres is $D_2 > D_1$. Since the porosity in our material tends to replace the binder phase, it will be represented by the intersection of $X_1^C$ either with $X_2$ (model A) or $X_2^C$ (model B) (Fig 5). If the two sets are independent, the covariance $C(h)$, which is the probability that the point doublet of size h is included in the porosity, is expressed by the product of the probabilities in $X_1^C$ and $X_2$ (or $X_2^C$) :

$$\text{Model A :} \quad C(h) = C_{X_1^C}(h)\, C_{X_2}(h)$$

$$(5)$$

$$\text{Model B :} \quad C(h) = C_{X_1^C}(h)\, C_{X_2^C}(h)$$

The volume fraction $V_V(X_1)$ of the first set is the volume fraction of grains $V_V(G)$ and is known from the analysis. The volume fraction $V_V(X_2)$ in the case of model A, or $V_V(X_2^C)$ in the case of model B, can be deduced from the measured porosity and from $V_V(G)$ : :

$$\text{Model A :} \quad V_V(P) = (1 - V_V(G))\, V_V(X_2)$$

$$(6)$$

$$\text{Model B :} \quad V_V(P) = (1 - V_V(G))\, V_V(X_2^C)$$

Hence, the experimental covariance function can be fitted by Eq. 5, where the model covariance functions are calculated by Eq. 2, 3 and 4, using measured values of the volume fractions and only adjusting the diameters $D_1$ and $D_2$ of the two sets. Least square fitting of the experimental data on the two models shows a rather good agreement for the two series analysed, a little better for model B (Fig 6). In addition, a model with only one boolean set is presented to show the necessity of a two-scale description of the microstructure.
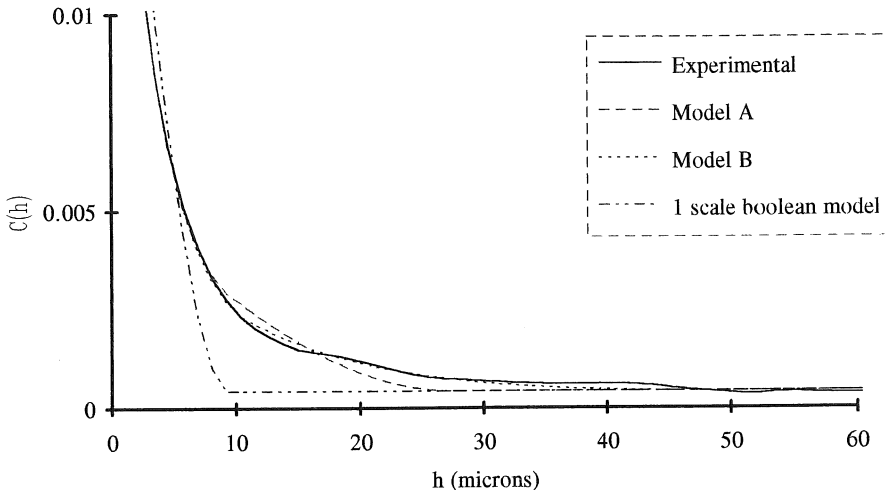


**Figure 6 :** Comparison of the experimental covariance function of porosity with those of the models (x 300)

Table 3 gives the diameters deduced from the models. Diameter $D_1$ is in the range 10-12 $\mu$m. The value calculated for a boolean set of volume fraction 0.74 and mean chord length 6.7 $\mu$m (measured values for $TiB_2$ grains) is 4.8 $\mu$m. This discrepancy shows the limits of the model, due probably to the assumption of spherical shape, and absence of size distribution at the two scales (porosity may occupy interstices between the largest $TiB_2$ grains, which could also explain the value of the mean chord length which is higher than for the binder phase, see Table 1). Determination of diameter $D_2$ is less precise : pores can be represented either as spherical 20-30 $\mu$m clusters or better as the complementary phase of 30-50 $\mu$m (grains+binder ) clusters, intersecting the complementary phase of grains.

**Table 3 :** Diameters deduced from the models (x 300)

| | Model A | | Model B | |
|---|---|---|---|---|
| | $D_1$ ($\mu$m) | $D_2$ ($\mu$m) | $D_1$ ($\mu$m) | $D_2$ ($\mu$m) |
| Series 1 | 9.6 | 27 | 11.2 | 45 |
| Series 2 | 10.1 | 22 | 11.9 | 32 |

## CONCLUSION

Analysis of chord length distributions and covariance functions allowed to characterize the microstructure of porosity in $TiB_2$-Fe composite materials. The size analysis showed that porosity tends to replace the binder phase in the microstructure. By representing the porosity as the intersection of two boolean sets, the experimental covariance function of this phase can be fitted pretty well, and the porosity represented either as 20-30 $\mu$m clusters, or as the complementary phase of 30-50 $\mu$m (grains+binder) clusters. Large scale fluctuations of the compositions are also evidenced, but their complete characterization would need analysis of a greater number of image series.

## AKNOWLEDGEMENTS

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