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CEN-VAMAS Round Robin on Grain Size Measurement for Advanced Technical Ceramics

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CEN-VAMAS Round Robin on Grain Size Measurement for Advanced Technical Ceramics - Final Report

by

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Summary

The results of a joint CEN/VAMAS Round Robin on grain size measurement for advanced technical ceramics have been analysed. The Round Robin was established primarily to verify the equivalence of line and circle methods for manual determination of mean linear intercept length proposed in a new CEN standard ENV 623-3, but also examined the possibilities of standardising measurements of porosity and grain size distribution using overlaid grid intersection counting methods. Two alumina ceramics with very different microstructures and a computer-drawn "ideal" microstructure were used. Twenty-five participants from Europe, USA and Japan took part. The results show that scatter levels of typically 20% between participants are obtained for measurement of the mean linear intercept length and the grain size distribution, and are primarily due to quality of specimen preparation, to selection of area for analysis, to interpretation of features observed and, to a lesser extent, also to random positioning of lines, circles and grids on the micrograph. These levels of scatter are considered to be broadly acceptable, bearing in mind the inherent randomness of microstructures. The results are considered to validate the equivalence of the line and circle methods for mean linear intercept size described in the new CEN standard. The 30% scatter for the grid method porosity measurement is mainly due to the low level of very fine porosity in the nominally dense ceramic used and also to the variable interpretation of a complicated micrograph, but is basically explainable by simple random counting variations. The grid methods for grain size distribution measurement seem suitable in principle for implementation in future standards. However, in all cases, the quality of micrograph preparation and interpretation needs to be given further attention because these factors appear to be of major importance in achieving consistent results between laboratories.

Keywords: grain size, intercept length, porosity, round robin, advanced technical ceramics

Contents

1. Introduction

Determination of the grain size of advanced technical ceramic materials is an important part of their characterization, both for material development and for quality control of commercially available materials. For this purpose it was agreed to formulate a standard within the framework of the European normalization programme for advanced technical ceramics.

A draft standard for manual grain size measurement has been prepared by CEN Technical Committee 184, Working Group 3 which essentially is based on the method described in ASTM E112, originally developed for use for metals. As such a standard for ceramic materials is proposed for the first time, it seemed useful to examine its applicability by means of a Round Robin exercise. The results of this Round Robin may give indications on the scatter of results between participants and the degree of difficulty experienced, leading to possible modification of the draft standard. Besides the results may resolve possible uncertainties about the range of applicability by the intended users, such as research or quality control laboratories.

This report gives an overview of the results of the Round Robin as run via CEN TC184/WG3 and VAMAS TWA3 from May to August 1992.

2. Objectives

The objectives of the Round Robin can be formulated as follows:

1 The primary objective of the Round Robin was to test the reproducibility of the CEN draft standard for grain size measurement by the manual determination of the mean linear intercept length. This was done in two ways:

   A The first way uses an "ideal" microstructure with no porosity and no second phase. This will determine the scatter of results between participants inherent in the random positioning of lines and circles on the "micrograph", which therefore represents the minimum attainable scatter without significant error due to misinterpretation of grain boundaries or other features.

   B The second way is more practical and is based on repeating the above procedure on a micrograph of a polished and thermally etched specimen as prepared by the organizers or by the participant. This will determine the additional scatter due to interpretation of a non-ideal microstructure and due to variation between areas randomly selected on specimens from a single batch of material. Two materials were used with strongly different grain size distributions, as the actual grain size distribution is likely to be of importance for the level of scatter.

2 The secondary objectives of the Round Robin were to test methods for use in possible future standards as they could be carried out at the same time on the same micrographs:

A To test a method for the determination of porosity

B To test a procedure for manually producing a grain size distribution.

In the draft CEN standard the use of automatic image analysis systems is not incorporated because of uncertainties in the methods employed and because comparisons between manual and automated methods were not available. The principal objectives of the Round Robin therefore deal with manual methods. To obtain information on the use of automatic methods, participants were asked to carry out the tasks with an automatic method if available such that a comparison between manual and automatic methods could be made.

3. Materials and methods

The participants of the round-robin (Appendix 0) have received the draft CEN standard (Appendix 1) and a set of instructions and materials (Appendix 2) to complete the tasks given in the instructions.

Part 1 of the round-robin deals with the determination of the mean intercept length by counting intersections of grains with a line or a circle as described in the draft standard. For this purpose a computer generated "microstructure" was used as given in Figure 3.1. This microstructure was generated as a pseudo-random 2-D Dirichlet tessellation containing 350 grains with no porosity.

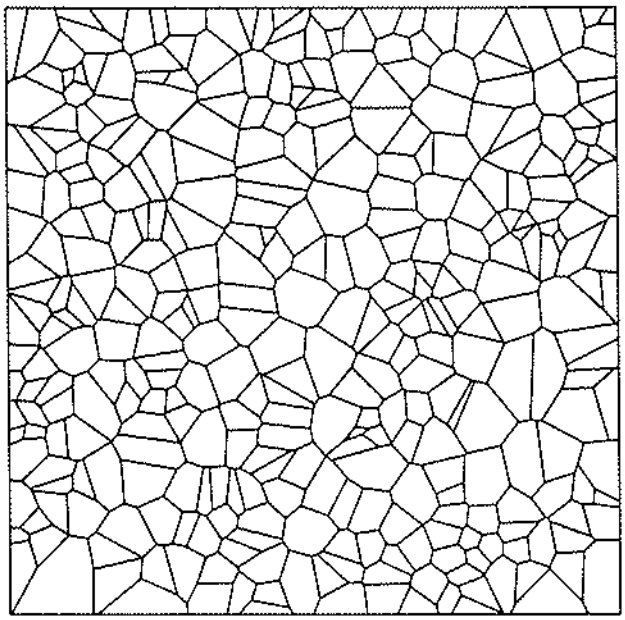

Figure 3.1 Computer drawn microstructure for Part 1, generated as a pseudo-random Dirichlet tessellation with 350 grains. True dimensions 159*159 mm, mean grain surface 72.2 mm^2.

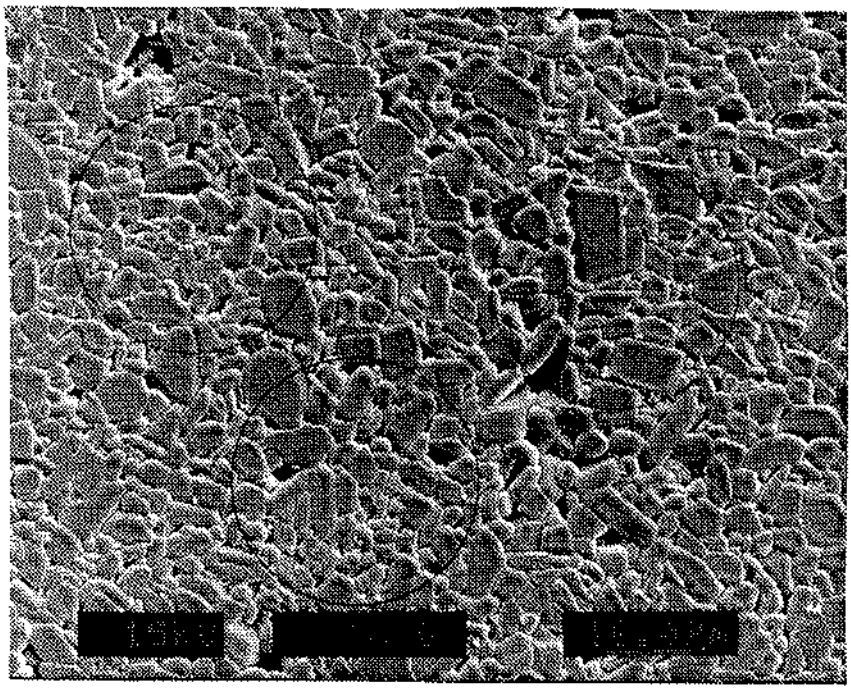

Figure 3.2 Micrograph of sample of material 1 from participant (scanning electron microscope).

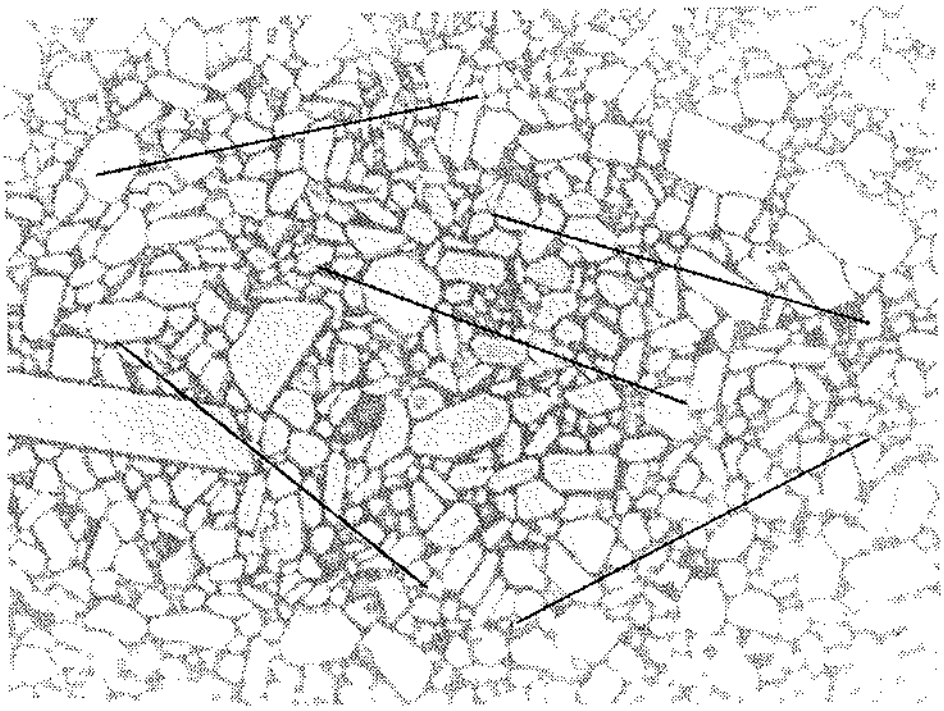

Figure 3.3 Micrograph of sample of material 1 from participant (optical microscope).

Part 2 of the Round Robin also deals with the determination of the mean intercept length by a line or circle method, but now using a micrograph made by the participant after polishing and etching of a supplied specimen of material 1 (96% brown alumina containing 4% submicrometre $MgTiO_3$ as second phase, sintered at about 1350 °C). Figures 3.2 and 3.3 show micrographs of this material as obtained by two of the participants.

Part 3 of the Round Robin asks the participants to use a micrograph of material 2 (99% alumina, sintered at about 1650 °C) (Figure 3.4) for measurement of the porosity with a supplied grid, for measurement of the mean intercept length by a line and circle method and for determination of the grain size distribution. This material has a wide range of grain size, but no obvious second phase, and was deliberately chosen to be a typical, but much more difficult case for microstructural analysis. The grid size, 5 mm, of the grid for porosity measurement (Figure 3.5) was obtained as a compromise between large enough for visual counting and small enough to be of the same scale as the features to be counted. Porosity can be determined by counting the number of intersections n_i of the grid that lie over a pore. If a grid intersection was perceived to lie on a pore boundary, this was to be counted as half an intersection. The ratio of n_i and the total number of intersections N, n_i/N, of the grid gives the porosity. For the determination of a grain size distribution using a grid method, the procedures in the annex of Appendix 2 were to be used. These methods essentially require that the distance between intercepts of grain boundaries and horizontal and vertical grid lines are measured.

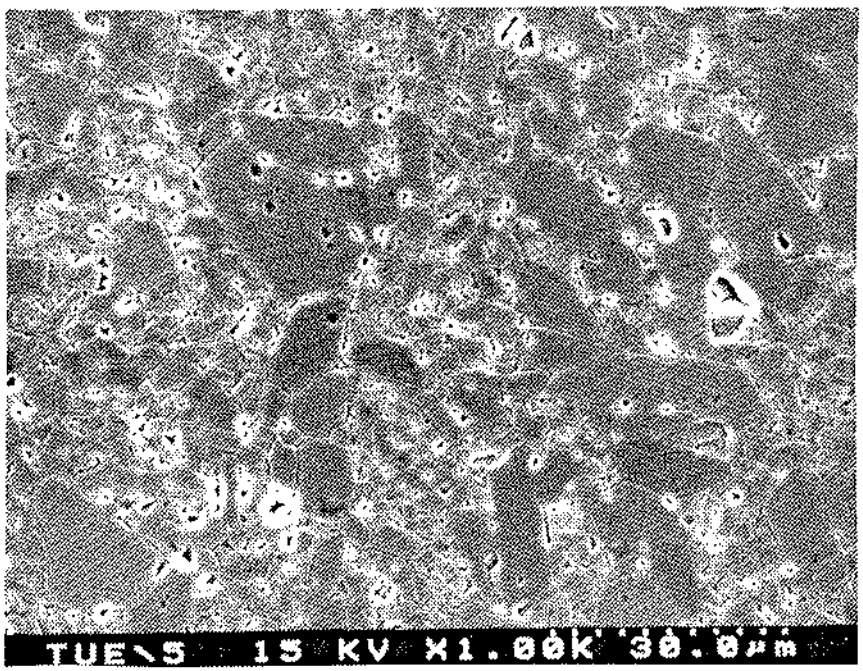

Figure 3.4 Micrograph of material 2 used for Part 3. True dimensions 242*176 mm (scanning electron microscope).

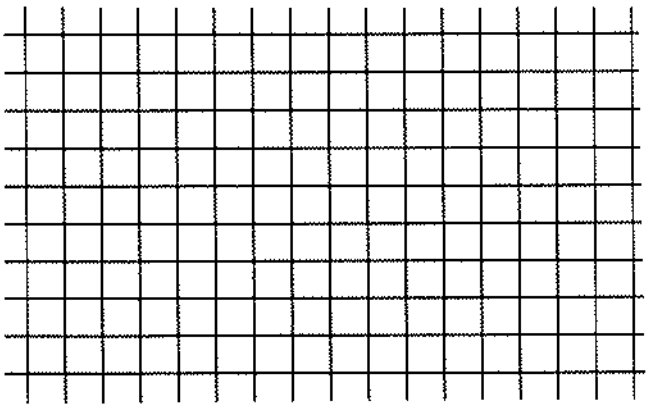

Figure 3.5 Grid used for porosity measurements with true grid size of 5 mm and line thickness of 0.15 mm.

Part 4 of the Round Robin essentially is the same as Part 3 except that now a specimen of material 2 was to be used by the participants to prepare a micrograph after polishing and etching. Figures 3.6 and 3.7 give examples of the micrographs returned by two of the participants.

The results of the various measurements were gathered on a reply form by each participant and returned for evaluation together with micrographs and figures made for the tasks.

At this point is is useful to give some formulae according to which certain statistical quantities were calculated and used in the evaluation of the results [1,2]. First, consider a set of N datapoints $\{x_1, x_2, x_3 \ldots x_N\}$. An estimate $\underline{x}$ for the mean μ is calculated from

$$\underline{x} = \frac{1}{N}\sum_{i=0}^{N} x_i$$

while an estimate s for the sample standard deviation σ is obtained from

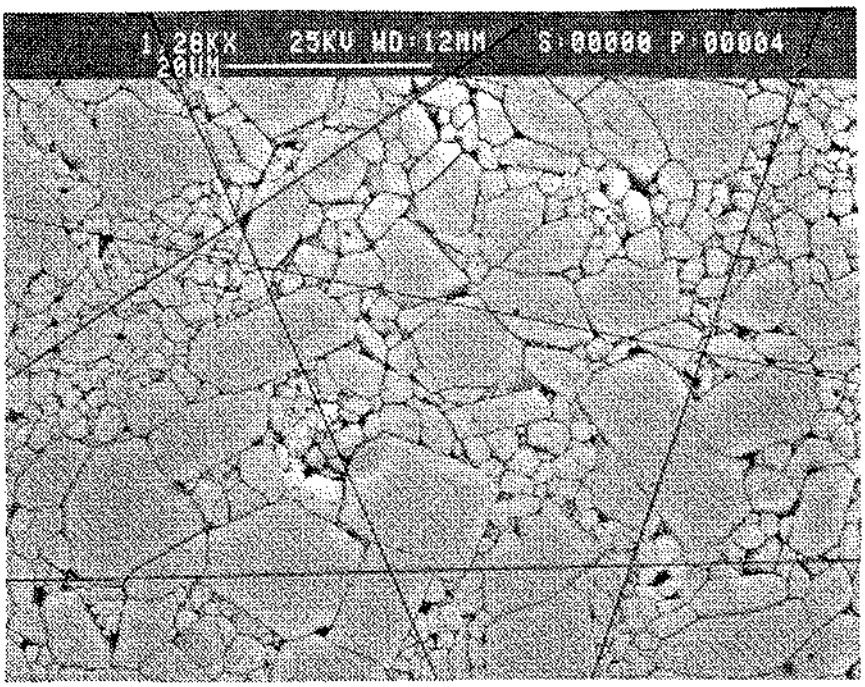

Figure 3.6 Micrograph of sample of material 2 from participant (scanning electron microscope).

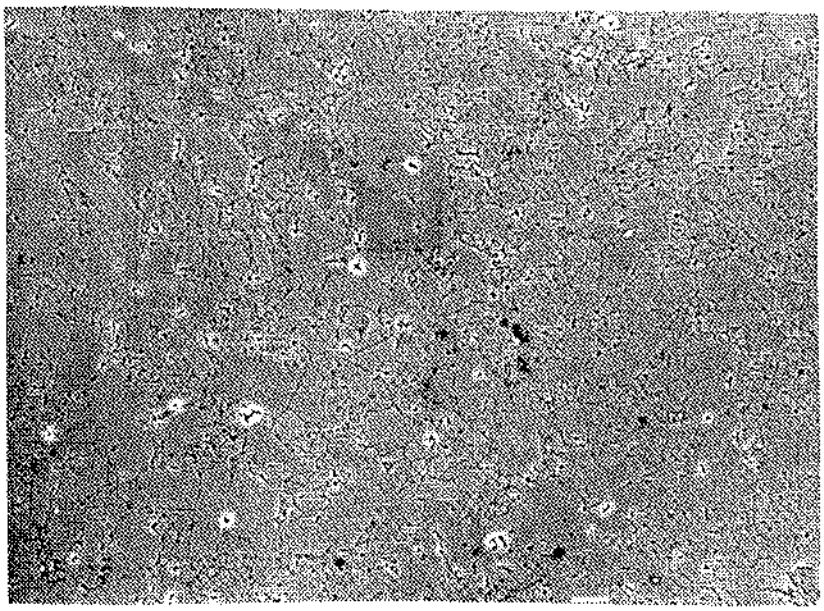

Figure 3.7 Micrograph of sample of material 2 from participant (scanning electron microscope).

$$s^2 = \sum_{i=1}^{N} \frac{(x_i - \underline{x})^2}{(N-1)}$$

To test whether two sets of data $\{x_1 .. x_N\}$ and $\{y_1 .. y_M\}$ have the same mean value, assuming that the data sets have a normal distribution and that the standard deviations are not known and not necessarily equal, use is made of the Welsh-Aspin T-test using the mean values $\underline{x}$ and $\underline{y}$ of the sets and the corresponding standard deviations s_x and s_y. The test statistic t is calculated as

$$t = \frac{|\underline{x} - \underline{y}|}{\sqrt{(}(s_x)^2/N + (s_y)^2/M)}$$

with an associated number of degrees of freedom n calculated from

$$n = \frac{((s_x)^2/N + (s_y)^2/M)^2}{((s_x)^2/N)^2/(N+1) + ((s_y)^2/M)^2/(M+1)} - 2$$

The hypothesis that $\underline{x}$ and $\underline{y}$ are identical will then be rejected if, for a two-tailed t-test with 95% confidence, the test statistic t having a t-distribution with n degrees of freedom, exceeds its critical value (to be found in textbooks, approximately 2 for n large enough).

4. Results

4.0 General

To identify the results of the 25 participants, a label Px was assigned to each participant with x ranging from 1 to 14, for participants from Europe and x ranging from 30 to 40 for participants from the USA and Japan. The labels were given according to the sequence in time in which the results reached the organizers. A list of participants is included in Appendix 0. The results of the participants have been taken as given on the reply form, except in those cases where an obvious calculation error was involved which was then corrected with approval of the participant in question.

4.1 Part 1: Computer drawn microstructure - Determination of mean linear intercept length

The results for Part 1 consist of values for the total line length (line method) or circle circumference (circle method) L, the total number of intersections n_i, and the mean linear intercept length l given by

$$l = L / n_i$$

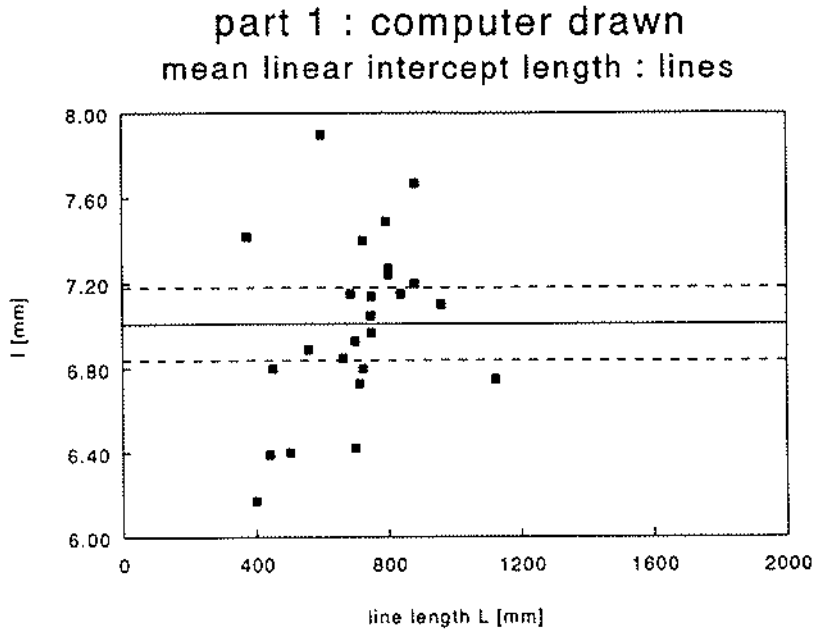

Figure 4.1.1 Mean linear intercept length l as a function of the line length L for all participants. ——— = average value for l, — — — = 95% confidence interval.

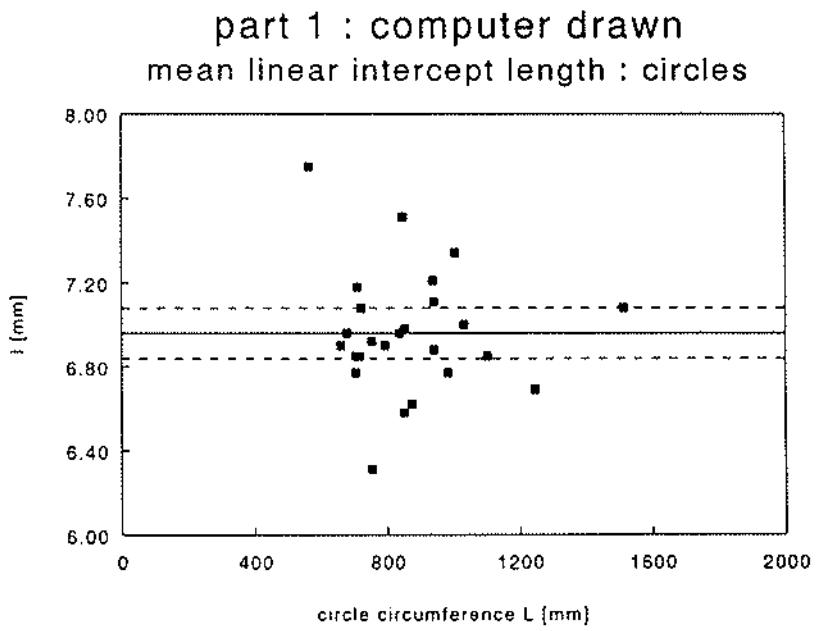

Figure 4.1.2 Mean linear intercept length l as a function of the line length L for all participants. ——— = average value for l, — — — = 95% confidence interval.

For both line and circle method the results are given in Table 4.1.1 and Figures 4.1.1 and 4.1.2, where the participants used 3 to 5 lines and circles for the analyses as proposed in the draft CEN standard.

Table 4.1.1 Results for computer drawn microstructure

(L = line length [mm], n_i = number of intersections, l = mean linear intercept length [mm])

	Lines			Circles		
Participant	L	n_i	l	L	n_i	l
P1	794.0	106.0	7.49	852.9	129.5	6.58
P2	837.0	117.0	7.15	1032.5	147.5	7.00
P3	1124.0	166.5	6.75	1515.1	214.0	7.08
P4	750.0	105.0	7.14	848.2	113.0	7.51
P5	503.0	78.5	6.40	659.7	95.5	6.90
P6	600.0	76.0	7.90	565.5	73.0	7.75
P7	800.0	110.0	7.27	942.5	132.5	7.11
P8	452.0	66.5	6.80	705.3	103.0	6.85
P9	960.0	136.0	7.10	754.0	109.0	6.92
P10	725.0	98.0	7.40	942.5	137.0	6.88
P11	747.5	106.0	7.05	838.8	120.5	6.96
P12	700.0	101.0	6.93	678.6	97.5	6.96
P13	880.0	123.0	7.20	1005.3	137.0	7.34
P14	375.0	50.5	7.42	722.6	102.0	7.08
P30	749.0	107.5	6.97	1247.2	86.5	6.69
P31	441.0	69.0	6.39	876.5	132.5	6.62
P32	713.0	106.0	6.73	716.3	104.5	6.85
P33	723.0	106.0	6.80	710.0	99.0	7.18
P34	401.0	65.0	6.17	703.7	104.0	6.77
P35	882.0	115.0	7.67	937.8	130.0	7.21
P36	686.0	96.0	7.15	1102.7	161.0	6.85
P37	664.0	97.0	6.85	984.9	145.5	6.77
P38	800.0	110.5	7.24	754.0	119.5	6.31
P39	558.0	81.0	6.89	854.5	122.5	6.98
P40	700.0	109.0	6.42	793.3	115.0	6.90

From Table 4.1.1 the following parameter values have been determined:

Lines

Average L for 25 participants	: 703 [mm]
Average n_i for 25 participants	: 100.1
Average l for 25 participants	: 7.01 [mm]
Sample standard deviation	: 0.41 [mm]
95% confidence interval for average l (t-distribution with 24 degrees of freedom)	: 6.84..7.18 [mm]

Circles

Average L for 25 participants	: 869 [mm]
Average n_i for 25 participants	: 125
Average l for 25 participants	: 6.96 [mm]
Sample standard deviation	: 0.29 [mm]
95% confidence interval for average l (t-distribution with 24 degrees of freedom)	: 6.84..7.08 [mm]

From these data, using the T-test on the average values for l, it is found that statistically the line and circle method do not yield a different mean linear intercept length (>95% confidence) and may therefore be considered identical. From Figures 4.1.1 and 4.1.2 is it also seen that in this Part of the Round Robin some results have been reported which clearly lie outside the 95% confidence intervals and therefore may need validation. Considering the values for n_i given in Table 4.1.1 one possible reason is obvious: most participants that used clearly less than 100 intersections yield a mean intercept length clearly outside the 95% confidence interval: for the line method P5, P6, P8, P14, P31, P34 and P39, and for the circle method P6. Therefore it may be concluded that at least 100 intersections are required to obtain a reasonable approximation for l, as is stated in draft CEN standard. The reason behind the relatively high value for the mean linear intercept length with the circle method reported by P4 may be that use was made of 3 concentric circles whereas in the draft CEN standard circles with random positioning of the centres are proposed. Use of concentric circles is believed to yield biased estimates, as shown by the result of P4. When these extreme values (P5, P6, P8, P14, P31, P34 and P39 for the line method and P4 and P6 for the circle method) are not taken into consideration one obtains:

Lines

Average L for 18 participants	: 791 [mm]
Average l for 18 participants	: 7.07 [mm]
Sample standard deviation	: 0.30 [mm]
95% confidence interval for average l (t-distribution with 17 degrees of freedom)	: 6.92..7.22 [mm]

Circles

Average L for 23 participants	: 884 [mm]
Average l for 23 participants	: 6.90 [mm]
Sample standard deviation	: 0.23 [mm]
95% confidence interval for average l (t-distribution with 22 degrees of freedom)	: 6.80..7.00 [mm]

Again using the T-test on the average value for l, with 95% confidence that the line and circle method yield the same result. The difference between the average values for l is therefore not significant, although it could be suspected that the influence of the 12% higher mean value for L for the circle method of 884 mm versus 791 mm for the line method has its influence as more grains are counted.

In summary it may be concluded that this Part of the round-robin served its purpose as its demonstrates that the line and circle methods for determination of the mean linear intercept length yield the same results. It also shows that the amount of scatter, defined as 2*standard deviation/mean value, between various participants is less than 10% provided at least 100 grain intersections with proper random positioning of lines and circles is used. This is considered as a reasonable value and may as a first indication be taken equal to $100\% / \sqrt{n_i}$. This scatter is likely to be due to the influence of random positioning of lines and circles on the micrograph as the influence of counting errors can be neglected. This scatter can also be interpreted as the minimum attainable scatter in these types of measurements as scatter due to misinterpretation of grain boundaries or other features is minimal.

4.2 Part 2: sample of material 1 - Determination of mean linear intercept length

The results for Part 2 also consist of values for the total line length (line method) or circle circumference (circle method) L with porosity excluded, the total number of intersections n_i and the mean intercept length l as determined from a micrograph prepared by each participant from a sample of material 1. The preparation procedures as reported by the participants are listed in Appendix 3.

For both line and circle method the results are given in Table 4.2.1 and Figures 4.2.1 and 4.2.2, where the values reported in millimetres have been divided by the magnification factor reported to obtain the values in micrometres. From Table 4.2.1 the following parameter values have been determined

Lines

Average L for 25 participants	: 323 [μm]
Average n_i for 25 participants	: 128
Average l for 25 participants	: 2.54 [μm]
Sample standard deviation	: 0.35 [μm]
95% confidence interval for average l (t-distribution with 24 degrees of freedom)	: 2.40..2.68 [μm]

Circles

Average L for 25 participants	: 376 [μm]
Average n_i for 25 participants	: 144
Average l for 25 participants	: 2.61 [μm]
Sample standard deviation	: 0.34 [μm]
95% confidence interval for average l (t-distribution with 24 degrees of freedom)	: 2.47..2.75 [μm]

Table 4.2.1 Results for sample of material 1.

(L = line length [μm], n_i = number of intersections, l = mean linear intercept length [μm])

	Lines			Circles		
Participant	L	n_i	l	L	n_i	l
P1	533.9	188.0	2.84	531.8	189.0	2.82
P2	270.3	109.0	2.48	353.2	133.0	2.66
P3	506.4	172.0	2.94	737.7	252.5	2.92
P4	278.5	105.0	2.65	350.3	109.0	3.21
P5	301.5	136.5	2.20	265.2	103.5	2.56
P6	256.6	95.0	2.70	241.9	78.0	3.10
P7	229.1	105.0	2.18	299.2	120.0	2.49
P8	284.4	111.5	2.55	293.7	121.5	2.42
P9	315.9	137.0	2.31	461.9	211.0	2.19
P10	277.0	113.5	2.44	311.5	148.0	2.10
P11	352.4	132.0	2.67	431.4	143.0	3.02
P12	325.9	140.0	2.33	463.7	183.0	2.53
P13	250.7	111.5	2.25	356.5	152.0	2.34
P14	250.0	71.0	3.52	377.0	113.5	3.32
P30	271.0	110.5	2.45	423.0	162.0	2.61
P31	295.3	141.0	2.09	292.0	128.0	2.28
P32	450.6	187.0	2.40	439.0	187.0	2.35
P33	480.0	157.0	3.05	360.5	130.0	2.77
P34	347.4	152.5	2.28	442.4	157.5	2.81
P35	499.3	168.0	2.97	391.6	138.0	2.84
P36	280.5	137.0	2.05	219.0	104.0	2.11
P37	375.6	148.0	2.54	528.0	203.0	2.60
P38	227.3	103.5	2.20	342.7	153.5	2.23
P39	145.2	51.5	2.82	245.9	91.5	2.69
P40	273.5	108.0	2.53	236.3	100.0	2.36

From these data it is readily found that also in this case the line and circle methods do not yield a statistically different mean linear intercept length (>95% confidence) and may therefore be considered identical. From the results given above it is seen that the scatter in the results is about 25% which is 15% higher than for Part 1 of the Round Robin. The reason for this not so obvious as only 3 participants used less than 100 grain intersections (P6, P14 and P39), which seems to play a disturbing role as discussed in Section 4.1. Again, the more extreme result of P4 for the circle method may be due to the use of concentric circles.

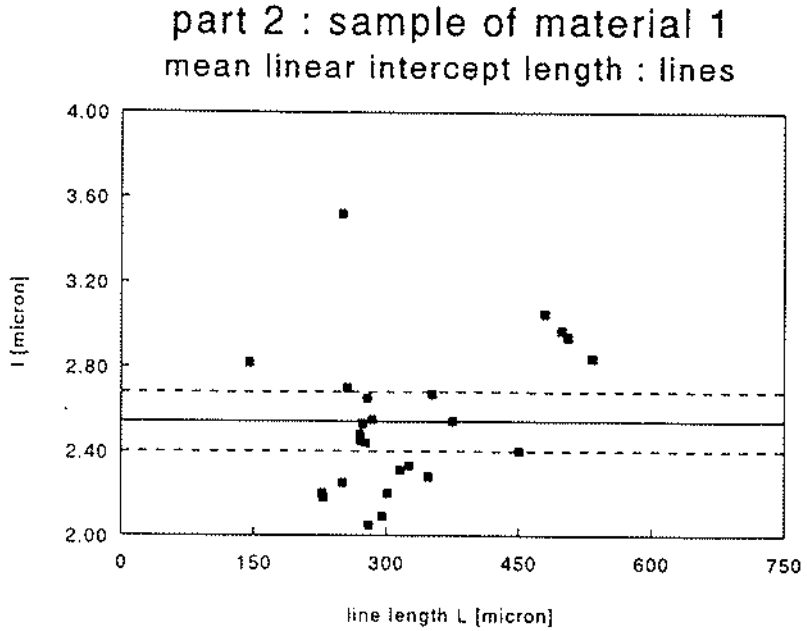

Figure 4.2.1 Mean linear intercept length l as a function of the line length L for all participants. ——— = average value for l, — — — = 95% confidence interval.

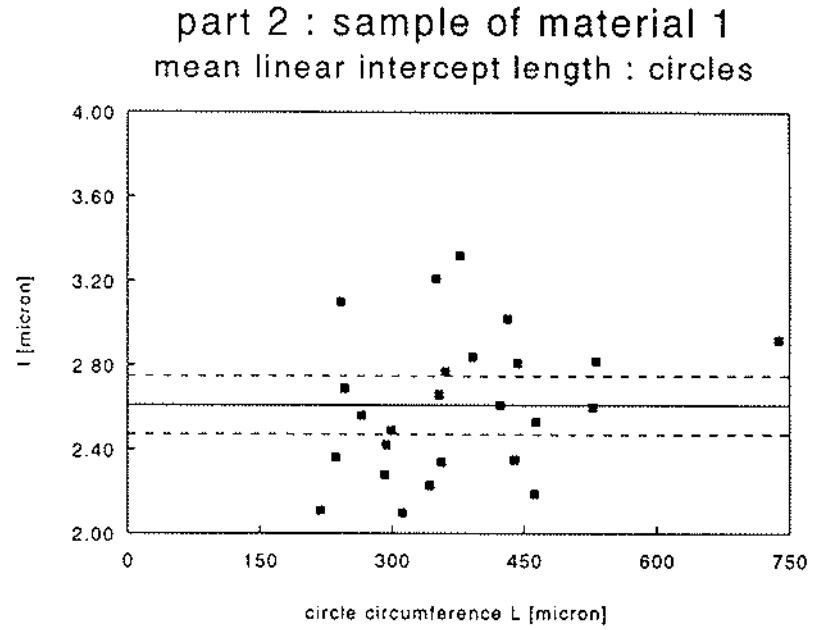

Figure 4.2.2 Mean linear intercept length l as a function of the circle circumference L for all participants. ——— = average value for l, — — — = 95% confidence interval.

It might be therefore be expected that this increase in scatter is due to a combination of variations in preparation techniques, variations in grain size between the distributed specimens (although taken from one batch), and different interpretation of features on a more complex micrograph. However, some systematic influence can be found if the etching temperatures and times, as listed in Appendix 3 are taken into consideration as listed in Table 4.2.2. Note that if a particular combination was used more than once the average result is given. The results of P4 for the circle method and of P6, P14 and P39 for the line and circle method were excluded.

Table 4.2.2 Mean linear intercept length values [μm] ranked with increasing etching temperature and time

Participant	Temperature[°C]	Time[min]	l, lines	l, circles
P31	1100	30	2.09	2.28
P36	1200	20	2.05	2.11
P9/P10	1200	30	2.38	2.15
P32	1250	15	2.40	2.35
P11	1250	60	2.67	3.02
P5	1300	30	2.20	2.56
P7	1350	5	2.18	2.49
P37	1350	6	2.54	2.60
P1/P8/P12	1350	10	2.57	2.59
P13	1350	15	2.25	2.34
P2/P4/P30/ P33/P34/P35/ P38/P40	1350	30	2.58	2.61
P3	1400	30	2.94	2.92

From Table 4.2.2 it can be deduced (with some caution because of the limited amount of data for certain combinations) that the mean intercept length increases with increasing etching temperature and time. This observation becomes rather clear when considering the values for the line method for 30 min etching where etching temperatures of 1100, 1200, 1300, 1350 and 1400 °C yield an averaged intercept length l of 2.09, 2.38, 2.20, 2.58 and 2.94 μm, respectively. From these data it might be concluded that etching temperature and time play a disturbing role. This need not necessarily be due to grain growth, although given a sintering temperature of about 1350 °C this cannot be excluded. The organisers have tried to analyse the data available to see if they can be described by means of an Arrhenius type of equation. It turns out that this is not possible because of the scatter in the results. A too strong etching procedure also may result in changes in the shape of the grains, e.g. rounding of corners and edges. This could be observed on many of the micrographs etched at higher temperatures, and could also result in the asking of small grains. It is difficult to estimate the precise influence of these factors on the scatter found because the results of the 8 specimen etched at 1350 °C, 30 min also gave a scatter of

25% (sample standard deviation approximatively 0.3 μm).

Some other comments on the results must also be made. The size of the micrographs made by the participants varied between approximatively 120*100 and 240*170 mm. No systematic influence of the size of micrographs could be found however. Most of the micrographs were made using a Scanning Electron Microscope (SEM), but 3 participants used an optical microscope (OM) namely P14, P31 and P37. Whether this has an influence cannot be determined. The results of P31 may be smaller than the average because of the low etching temperature, whereas in the eyes of the organisers the micrograph of P37 is rather unsharp. It is important to note that the OM micrographs appear quite different from those taken by SEM in the sense that grain boundaries are less pronounced, possibly because of light scattering introduced by the grains, which does not occur in the SEM micrographs. This might lead to an alternative interpretation of the micrograph. Some of the scatter in the results may also be due to the fact that the micrographs made by SEM have not been made with the same set-up of the SEM. Marked differences in voltage and working distance are recorded, although no systematic influence could be found. However, these differences may lead to a different interpretation of the microstructure as grain boundaries etc may be more or less pronounced on the micrograph. The applied polishing/etching procedure is therefore likely to influence the results as it may influence the set-up for the SEM/OM required to produce a micrograph which is acceptable in the eyes of the operator.

At this moment it can be stated that in principle the methods for mean linear intercept length measurement agree well and that the main causes for the scatter in the results are

- the influence of random positioning of lines and circles (as analysed in Part 1);
- the influence of making measurements on different prepared areas;
- the influence of etching temperature and time through possible grain growth and change of shape of the grains;
- the influence of the polishing/etching procedure and the set-up of the SEM or OM to make a micrograph where a certain amount of subjectivity is introduced.

The latter two causes only are related to micrograph preparation and may obscure whether additional scatter is introduced due to the interpretation of a more complicated micrograph than the one used in Part 1. On the other hand it can be stated that material 1 has a "nice" microstructure (equiaxed grains of relatively uniform size with clearly marked grain boundaries) and does not easily give rise to grain pluck-out so that the interpretation as such should not have been too complicated and should not have led to substantial additional scatter. Therefore the 15% increase in scatter could be attributed to the influence of micrograph preparation only.

4.3 Part 3 : Micrograph of material 2

Part 3 of the Round Robin uses a micrograph of material 2 as prepared by the organisers (see chapter 3, Figure 3.4). This micrograph was used for measurement of porosity, mean linear intercept length and grain size distribution, as will be discussed in the next Sections.

4.3.1 Porosity measurement

The results for the porosity measurement consist of the number of grid intersections N used for counting pores, the number of pores n counted. The porosity is then determined by the ratio n/N, expressed in % for convenience. The results given by the participants are shown in Table 4.3.1.1 and Figure 4.3.1.1

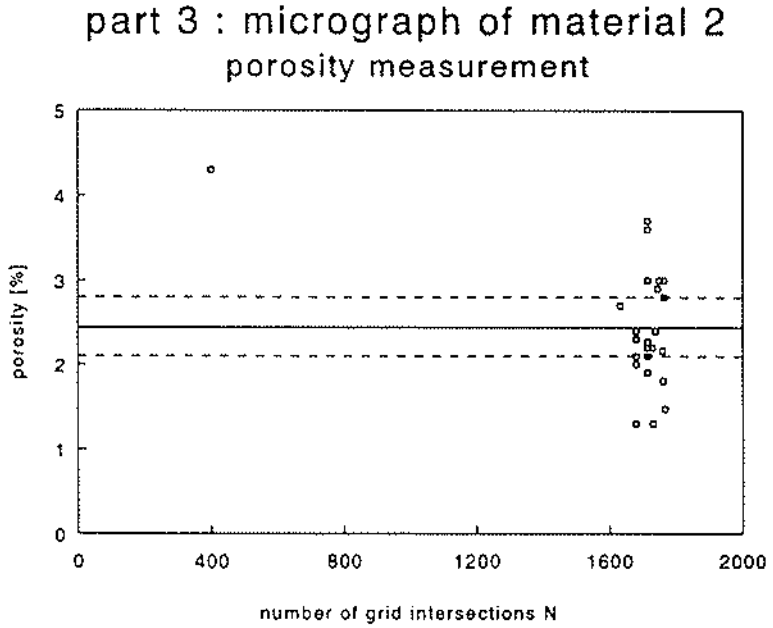

Figure 4.3.1.1 Porosity as a function of the number of grid intersections N for all participants. ——— = average value for porosity, — — — = 95% confidence interval.

Table 4.3.1.1 Results for porosity measurement

(N = number of grid intersections, n = number of pores, porosity in %)

Participant	N	n	porosity
P1	1728	38.0	2.2
P2	1680	38.0	2.3
P3	1680	40.5	2.4
P4	1680	36.0	2.1
P5	1632	43.5	2.7
P6	400	17.0	4.3
P7	1715	64.0	3.7
P8	1749	52.5	3.0
P9	1680	33.0	2.0
P10	1715	32.5	1.9
P11	1680	21.5	1.3
P12	1715	38.0	2.2
P13	1715	61.0	3.6
P14	1715	37.0	2.2
P30	1745	50.0	2.9
P31	1764	49.0	2.8
P32	1731	23.5	1.3
P33	1715	51.5	3.0
P34	1766	26.0	1.5
P35	1715	39.0	2.3
P36	1758	38.0	2.2
P37	1715	36.5	2.1
P38	1764	53.0	3.0
P39	1738	42.0	2.4
P40	1761	31.0	1.8

From Table 4.3.1.1 the following parameter values have been determined:

Average N for 25 participants	: 1666
Average n for 25 participants	: 40
Average porosity for 25 participants	: 2.4 [%]
Sample standard deviation	: 0.7 [%]
95% confidence interval for porosity (t-distribution with 24 degrees of freedom)	: 2.1..2.8 [%]

The scatter in the values for the porosity is about 60% which may seem considerable. However one has to recognize that the porosity as such is a relatively small number and therefore error-prone. Part of the scatter may be attributed to difficulties in counting because the large number of grid intersections to be examined probably causes eyestrain. The positioning of the grid on the micrograph and the interpretation of what is a pore are

likely to have the biggest influence, however. A finer grid might be appropriate but will be more time consuming and therefore not useful. A flat unetched surface with no confusing relief would probably have been a better test.

4.3.2 Determination of mean linear intercept length

For the determination of the mean linear intercept length again the results consist of values for the total line length (line method) or circle circumference (circle method) L, the total number of intersections n_i and the mean intercept length l. The values are given in millimetres as measured from the micrograph. For both line and circle methods the results are given in Table 4.3.2.1 and Figures 4.3.2.1 and 4.3.2.2.

Table 4.3.2.1 Results for mean linear intercept length measurement.

(L = line length [mm], n_i = number of intersections, l = mean linear intercept length [mm])

	Lines			Circles		
Participant	L	n_i	l	L	n_i	l
P1	733.0	136.0	5.39	1017.9	214.0	4.75
P2	817.0	168.5	4.85	1131.0	235.5	4.8
P3	1102.0	210.0	5.25	2075.0	422.5	4.91
P4	900.0	162.0	5.56	914.0	160.0	5.71
P5	750.0	138.5	5.42	659.7	90.5	7.29
P6	600.0	75.0	8.00	565.5	80.0	7.07
P7	633.0	121.5	5.21	494.7	111.0	4.46
P8	679.0	114.0	5.96	859.5	146.0	5.89
P9	783.0	132.0	5.94	996.6	192.5	5.18
P10	728.0	121.5	6.00	937.0	196.0	4.78
P11	928.0	133.0	6.98	1140.4	208.0	5.48
P12	645.0	112.0	5.76	915.0	178.5	5.13
P13	700.0	121.5	5.76	1193.5	207.0	5.76
P14	375.0	74.5	5.03	659.7	110.5	5.97
P30	1020.0	180.0	5.67			
P31	585.0	124.0	4.70	876.5	177.5	4.94
P32	790.0	134.0	5.89	1008.4	194.5	5.18
P33	768.0	151.5	5.07	667.0	131.5	5.07
P34	533.5	82.5	6.47	861.5	174.0	4.95
P35	1061.0	175.5	6.04	937.7	138.0	6.79
P36	1042.0	216.5	4.81	1181.2	222.5	5.31
P37	910.5	150.0	6.07	907.6	162.5	5.59
P38	600.0	103.0	5.83	550.0	102.5	5.38
P39	523.0	86.5	6.00	1057.1	171.0	6.18
P40	750.0	155.0	4.84	663.0	131.0	5.06

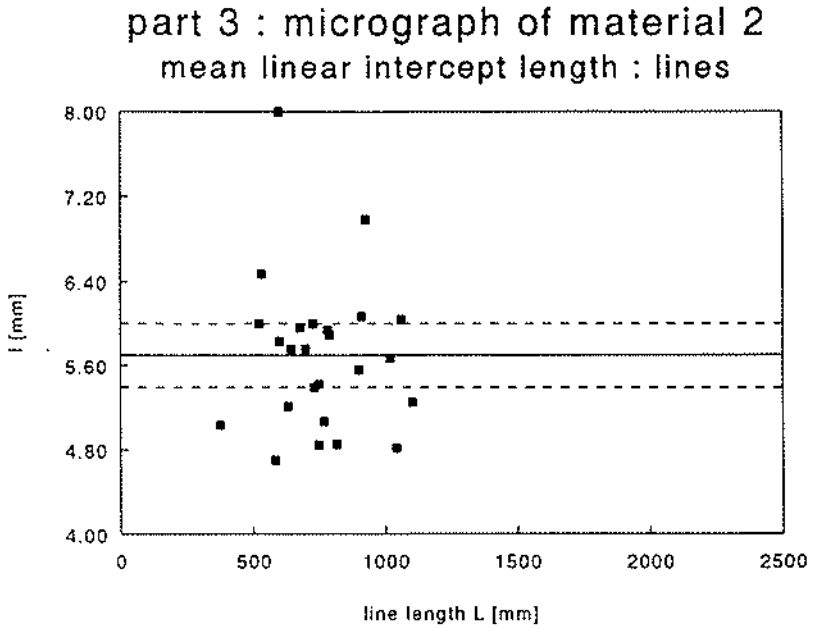

Figure 4.3.2.1 Mean linear intercept length l as a function of the line length L for all participants. ——— = average value for l, — — — = 95% confidence interval.

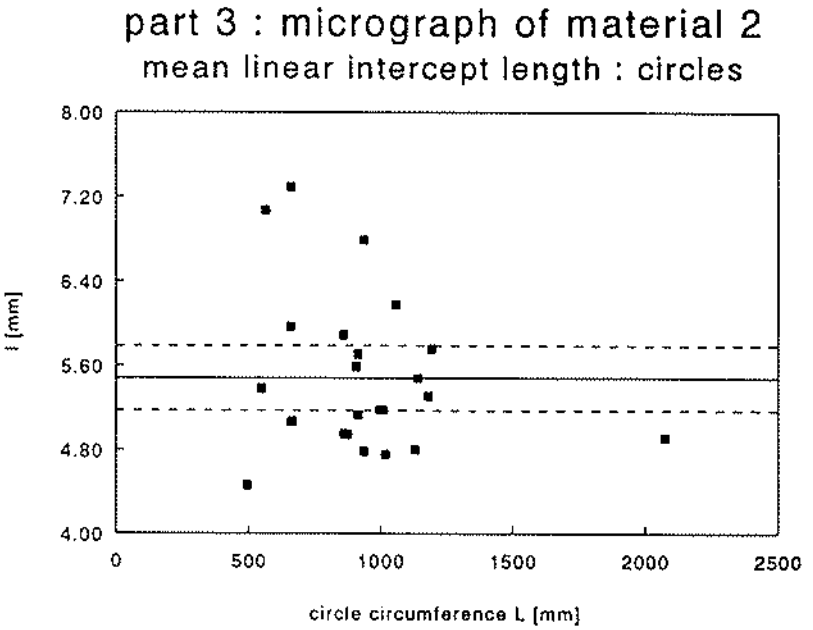

Figure 4.3.2.2 Mean linear intercept length l as a function of the circle circumference L for all participants. ——— = average value for l, — — — = 95% confidence interval

From Table 4.3.2.1 the following parameter values have been determined:

Lines

Average L for 25 participants	: 758 [mm]
Average n_i for 25 participants	: 135
Average l for 25 participants	: 5.70 [mm]
Sample standard deviation	: 0.73 [mm]
95% confidence interval for average l (t-distribution with 24 degrees of freedom)	: 5.39..6.00 [mm]

Circles

Average L for 24 participants	: 928 [mm]
Average n_i for 24 participants	: 173
Average l for 24 participants	: 5.48 [mm]
Sample standard deviation	: 0.75 [mm]
95% confidence interval for average l (t-distribution with 23 degrees of freedom)	: 5.17..5.79 [mm]

From these data is readily found that also in this case the line and circle methods do not yield a statistically different mean linear intercept lengths (>95% confidence). As was concluded in Part 1 the results from cases in which less than 100 grain intersections are counted are likely to be biased. This would explain why for the line method the results of P6, P14, P34 and P39 and for the circle method the results of P5 and P6 are more extreme. Also, the result of P4 for the circle method may be doubtful as concentric circles were used which, as discussed in Part 1 and Part 2, is likely to yield biased results. Excluding these results the average values for l become 5.57 and 5.31 mm for the line and circle method, respectively, with corresponding standard deviations of 0.55 and 0.55 mm which gives rise to 95% confidence intervals of 5.32..5.82 and 5.06..5.56 [mm] respectively. Again use of the T-test on the average values for l shows that with 95% confidence the line and circle methods yield the same value. With some of the results excluded, the scatter between the participants is reduced from about 30% to about 20% which is higher than for Part 1 and 5% less than for Part 2. In this case all participants used the same micrograph, such that the disturbing influences of specimen preparation can be neglected, the increase in scatter with respect to Part 1 and Part 2 is likely to be due to:

- the influence of a microstructure with a wider grain size distribution as used in Part 2:

- increased difficulties in evaluation of a "real" micrograph from a material with a more complicated microstructure where misinterpretation of grain boundaries and other features is possible.

The latter reason would also explain the increase in scatter with respect to Part 2 (if the larger Part of the scatter in Part 2 is devoted to the influence of micrograph preparation) as the microstructure of material 1 is "nicer" and therefore less liable to misinterpretation.

4.3.3 Determination of grain size distribution

As discussed in Section 2, one of the targets of the Round Robin is to test a method for determination of a grain size distribution. For this purpose two methods were proposed as given in Appendix 2. The results of the analyses by the participants were returned on a floppy disk and subsequently processed. As a first check on the results the average l and the standard deviation s of the grain size distribution returned by each participant, will be discussed. Table 4.3.3.1 and Figures 4.3.3.1 and 4.3.3.2 show the data obtained.

Table 4.3.3.1 Results for grain size distribution measurement.

(N = number of grid intersections, l = mean intercept size [mm], s = standard deviation of intercept sizes [mm])

	Method 1			Method 2		
Participant	N	l	s	N	l	s
P1	558	5.49	6.54			
P2	464	6.11	5.74			
P3	197	5.59	5.87			
P4	214	5.67	5.94			
P5	426	6.16	5.57	510	6.32	5.88
P7	152	5.19	5.14	302	4.48	4.83
P8	607	6.87	5.90			
P9	540	5.40	5.70	618	5.40	5.50
P10	451	4.80	5.20	736	4.80	5.60
P11	489	6.35	5.97			
P12	432	5.46	5.20			
P13	362	5.36	5.04	512	5.67	5.64
P14	433	5.51	5.17	574	5.72	5.69
P30	412	6.40	6.03			
P32	316	6.03	6.23	579	5.65	5.54
P33	331	5.20	5.50	403	5.20	5.40
P34	359	5.70	5.70			
P36	571	5.30	5.77	716	5.35	5.85
P37	497	4.80	5.49			
P38	188	7.09	5.88	218	7.19	5.86
P39	210	7.20	6.10			
P40	668	5.32	5.49			

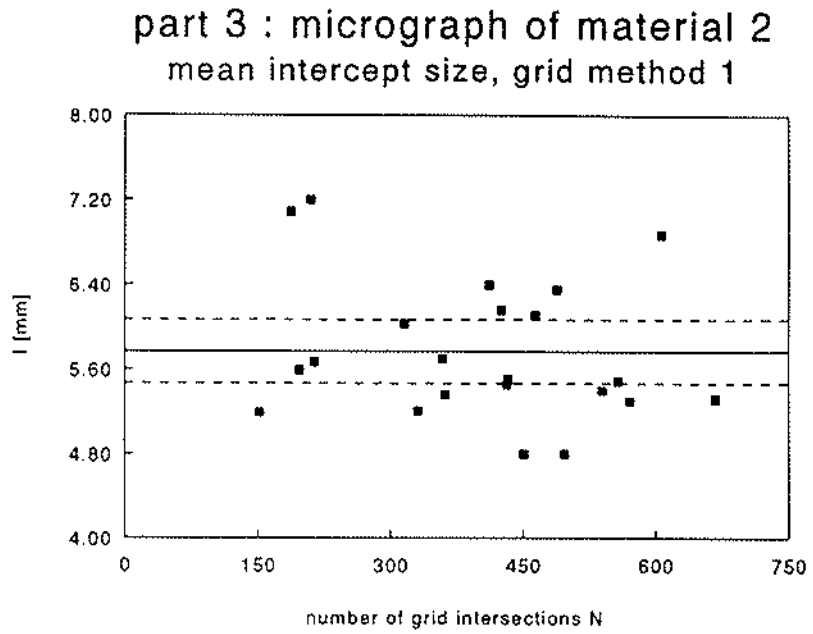

Figure 4.3.3.1 Mean intercept size l as a function of the number of grid intersections N for all participants for Method 1. ——— = average value for l, — — — = 95% confidence interval.

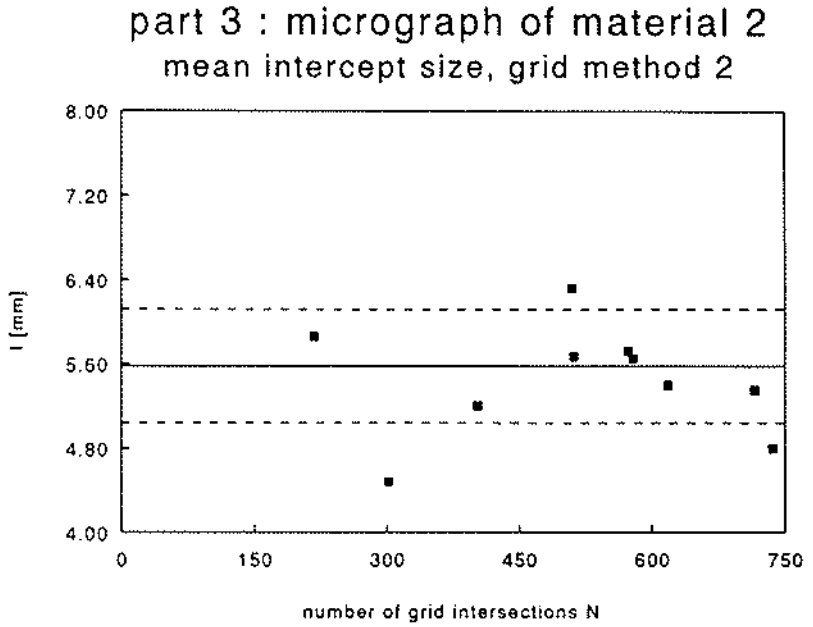

Figure 4.3.3.2 Mean intercept size l as a function of the number of grid intersections N for all participants for Method 2. ——— = average value for l, — — — = 95% confidence interval.

From Table 4.3.3.1 the following parameter values have been determined:

Method 1

Average N for 22 participants	: 404
Average l for 22 participants	: 5.77 [mm]
Sample standard deviation	: 0.68 [mm]
95% confidence interval for average l (t-distribution with 21 degrees of freedom)	: 5.47..6.07 [mm]

Method 2

Average N for 10 participants	: 517
Average l for 10 participants	: 5.58 [mm]
Sample standard deviation	: 0.76 [mm]
95% confidence interval for average l (t-distribution with 9 degrees of freedom)	: 5.04..6.12 [mm]

Using the 95% confidence intervals for the mean linear intercept length determined on the same micrograph with the line and circle methods (Section 4.3.2) of 5.39..6.00 and 5.17..5.79 [mm] respectively, it is readily seen that the values for the average l given above are well within these confidence intervals. This indicates that the mean value of the grain size distributions determined agrees well with the one obtained with the line and circle method, as it should be. Also the scatter in l is of the same order of magnitude (25%) as was found for the line and circles methods. It is worthwhile mentioning that most of the participants used a grid of 15*15 to 20*20 mm. Participant P8 used a grid of 10*10 mm and shows a rather large value for l, whereas participants P7 and P33 used a grid of 60*40 and 28*28 mm respectively and obtained a rather small value for l. This dependence on the grid size is not unexpected as for a small grid, large grains are likely to be intersected more often and therefore the mean grain size will increase. In the case of a coarse grid the question arises as to whether sufficient intersections are present to have reliable statistics as can be concluded from the results of P38 and P39 who used a grid size of 30*30 and 27*27 mm respectively. This question cannot be answered at this moment as from Table 4.3.3.1 and Fig. 4.3.3.3 and 4.3.3.4 no obvious relation between number of intersections and grain size is found. If the results of P7, P8 and P33 are excluded the average value for l for Method 1 and Method 2 becomes 5.78 and 5.76 [mm], respectively with a corresponding standard deviation of 0.64 and 0.67 [mm]. These results are not significantly different from those derived from all results in Table 4.3.3.1. The grid dimensions of 15*15 to 20*20 mm are such that in general a grid of about 10*10 lines could be drawn on the micrograph as asked in the instructions: "select the spacing of the grid to be approximately the same size as the largest grains so that each of these are not normally counted twice". Here it was not meant that one should select the single largest grain on the micrograph, but a "typical" large grain which is about 20 mm in size. The word "approximately" was used deliberately by the organisers to see how participants reacted to a situation which contained conflicts, but obviously such an expression has to be used with caution. Some of the participants did take the largest grain on the micrograph and could therefore not draw a grid with at least 10 lines. The requirement for a 10*10 grid was given by the need to have sufficient grid intersections for statistical reliability and seems to have been sufficient.

No obvious difference between Method 1 and Method 2 is observed, although the number of results for Method 2 is too small to give a definitive answer. The differences between these methods expected to occur are therefore likely to be of lesser importance in practice than other factors, such as the influence of random positioning of the grid, the grid size and uniformity of the microstructure, which appear to have a larger impact on the variability of the results.

Next the actual grain size distribution will be discussed in more detail. For this purpose the data returned by each participant were sorted in ascending order such that the cumulative and relative frequency distribution functions could be computed. The number of classes was set to 10 with a equidistant logarithmic spacing between the minimum and maximum value found. As an example, for some of the participants Figure 4.3.3.3 shows the cumulative frequency distributions as a function of the class middle value x for Method 1, whereas Figure 4.3.3.4 shows the corresponding relative frequency distributions.

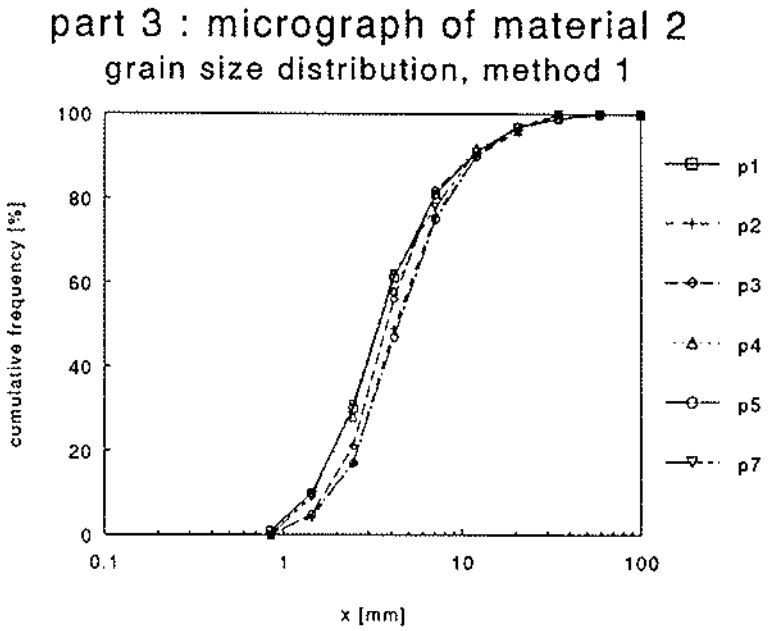

Figure 4.3.3.3 Cumulative frequency distribution for grain size determined with Method 1.

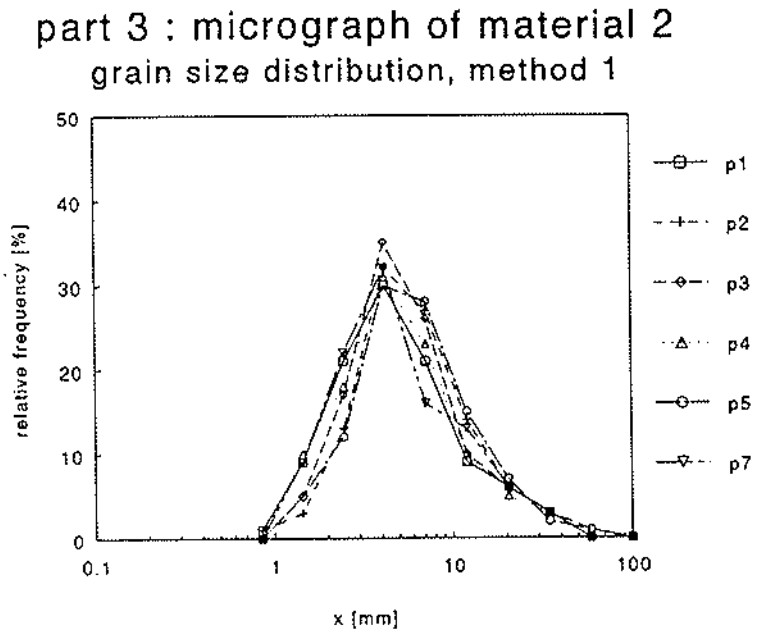

Figure 4.3.3.4 Relative frequency distribution for grain size determined with Method 1.

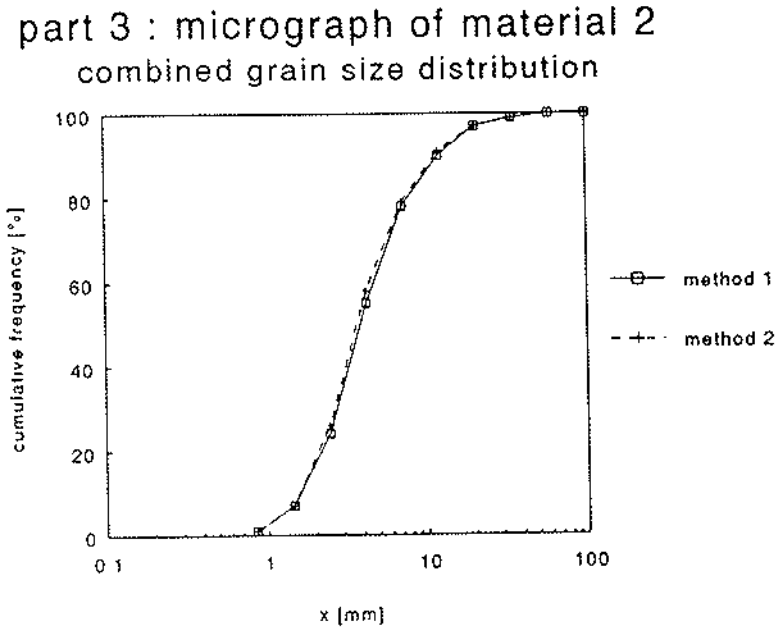

Figure 4.3.3.5 Combined cumulative frequency distribution for grain sizes determined with Method 1 and Method 2.

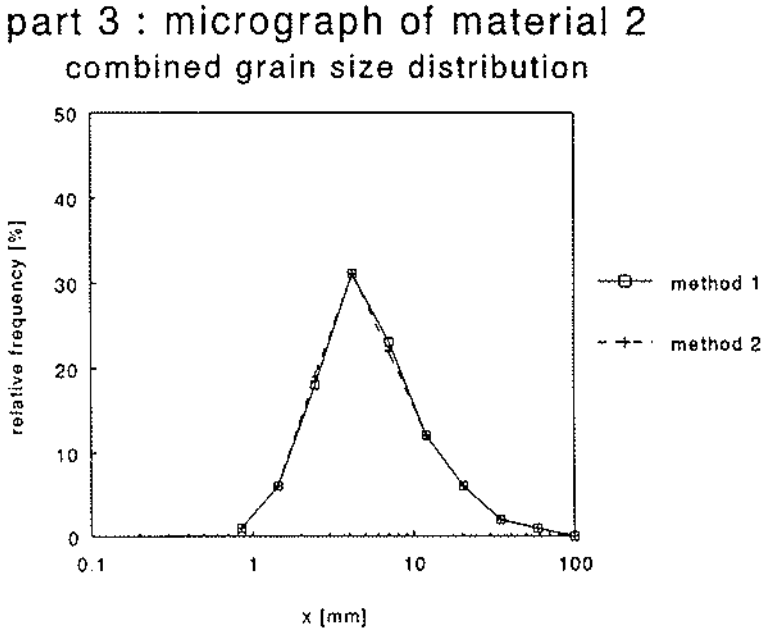

Figure 4.3.3.6 Combined relative frequency distribution for grain sizes determined with Method 1 and Method 2.

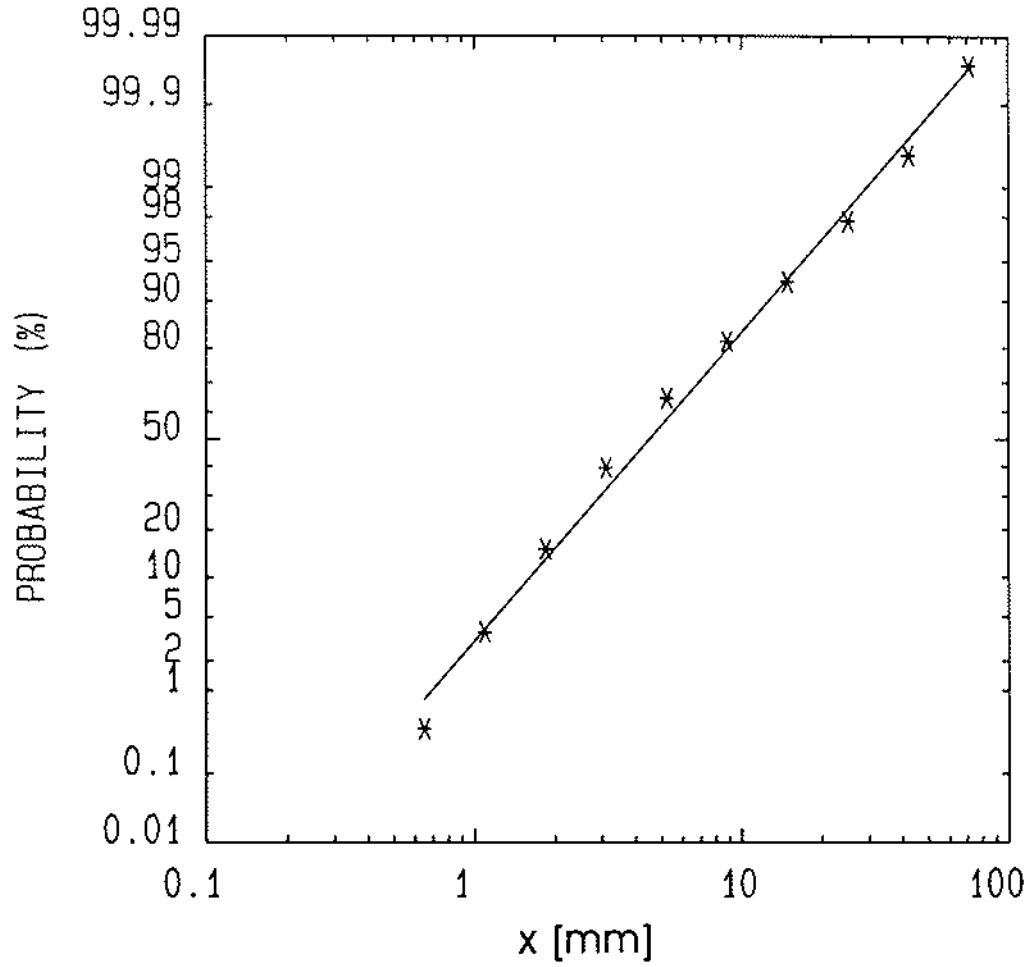

Figure 4.3.3.7 Example of a cumulative frequency distribution plotted on a log-normal scale. Straight line represents least squares fit.

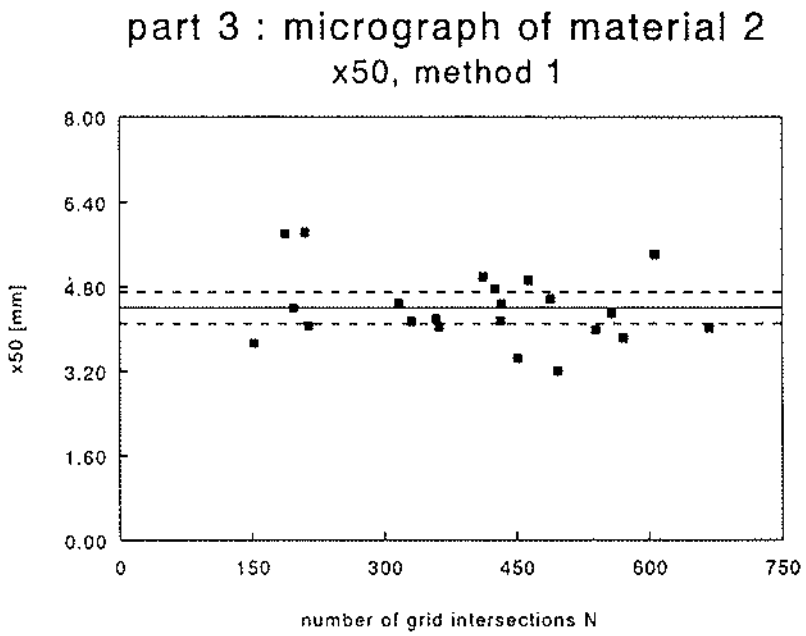

Figure 4.3.3.8 Computed values for 50% probability grain size x_{50} for Method 1. ——— = average value for x_{50}, — — — = 95% confidence interval.

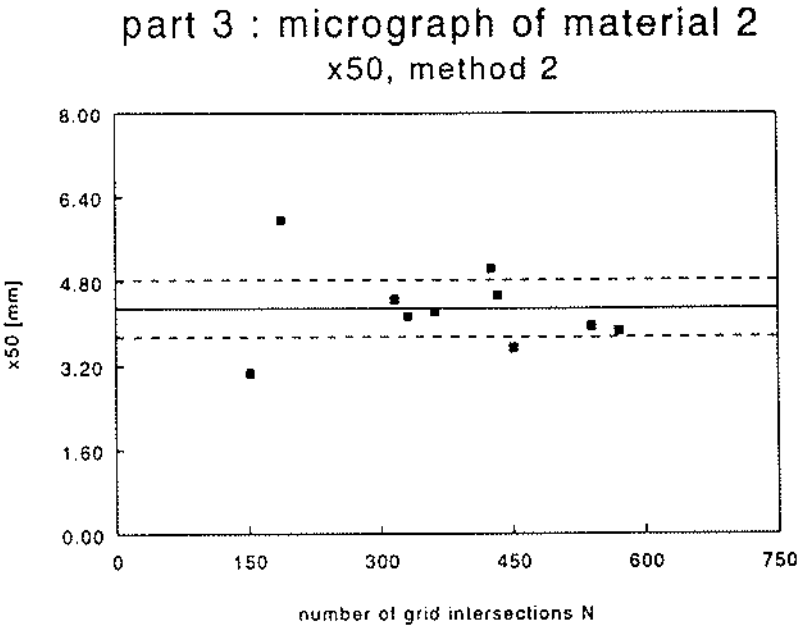

Figure 4.3.3.9 Computed values for 50% probability grain size x_{50} for Method 2. ——— = average value for x_{50}, — — — = 95% confidence interval.

The values for the grain sizes reported by all participants were also gathered in a single distribution calculated from the combined values measured. The cumulative and relative frequency distributions for Method 1 and Method 2 are shown in Figures 4.3.3.5 and 4.3.3.6.

For a quantitative description of these data, the data were plotted on a log-normal scale. An example is shown Fig. 4.3.3.7.

Using a linear regression method, values for the slope and intercept were obtained from which the 50% probability grain size x_{50} and the standard deviation σ_{lnx} of the log-normal distribution easily can be calculated. The computed values are listed in Table 4.3.3.2, while the values of x_{50} for Method 1 and Method 2 are also given in Figs. 4.3.3.8 and 4.3.3.9.

Table 4.3.3.2 Computed values for 50% probability grain size x_{50} [mm] and standard deviation σ_{lnx}

Participant	Method 1		Method 2	
	x_{50}	σ_{lnx}	x_{50}	σ_{lnx}
P1	4.30	0.88		
P2	4.93	0.80		
P3	4.40	0.82		
P4	4.07	0.87		
P5	4.76	0.78	5.04	0.79
P7	3.73	0.88	3.06	0.88
P8	5.42	0.74		
P9	3.98	0.85	3.95	0.84
P10	3.44	0.84	3.54	0.87
P11	4.57	0.85		
P12	4.16	0.79		
P13	4.04	0.78	4.22	0.81
P14	4.48	0.78	4.48	0.81
P30	4.99	0.78		
P32	4.50	0.84	4.45	0.79
P33	4.15	0.80	4.14	0.80
P34	4.20	0.85		
P36	3.83	0.88	3.86	0.88
P37	3.20	0.95		
P38	5.81	0.67	5.96	0.67
P39	5.83	0.72		
P40	4.03	0.81		

From Table 4.3.3.2 the following mean values and standard deviations were obtained:

Method 1

Mean value for x_{50}	: 4.40 [mm]
Sample standard deviation	: 0.67 [mm]
95% confidence interval for average x_{50} (t-distribution with 21 degrees of freedom)	: 4.10 .. 4.70 [mm]

Method 2

Mean value for x_{50}	: 4.28 [mm]
Sample standard deviation	: 0.76 [mm]
95% confidence interval for average x_{50} (t-distribution with 9 degrees of freedom)	: 3.73 .. 4.82 [mm]

From these results it can be concluded that the lognormal distribution fits the data very well (correlation coefficient > 0.99). Also no deviations from a unimodal distribution can be found. The scatter in the values for x_{50} is about 30% as was found for the average grain size. Also it is observed that Method 1 and Method 2 do not yield significantly different results.

As a comparable analysis for grain size distribution measurement on ceramic materials is not available to the organizers knowledge, it is difficult to give an appreciation for this scatter. Given the consistency with the scatter in the mean linear intercept length and average grain size and the fact that the microstructure of this material is not ideal, it is felt that the procedures defined in this Round Robin for grain size distribution measurement are suitable for implementation in future standards.

4.4 Part 4 : Sample of material 2

Part 4 of the Round Robin uses a sample of Material 2 which was polished and etched by participants to produce a micrograph. The preparation procedures reported by the participants are listed in Appendix 4, from which it can be concluded that in most cases the same polishing procedure was adopted as for Material 1 (the etching conditions were generally different however). The resulting micrograph was used for measurement of mean linear intercept length and grain size distribution as will be discussed in the next Sections.

4.4.1 Determination of mean linear intercept length

For the determination of the mean linear intercept length once more the results consist of values for the total line length (line method) or circle circumference (circle method) L, the total number of intersections n_i and the mean intercept length l. The values are given in micrometres as measured from the micrographs. For both line and circle methods the results are given in Table 4.4.1.1 and Figures 4.4.1.1 and 4.4.1.2.

Table 4.4.1.1 Results for mean linear intercept measurement

(L = line length [μm], n_i = number of intersections, l = mean linear intercept length [μm])

Participant	Lines			Circles		
	L	n_i	l	L	n_i	l
P1	301.8	121.0	2.49	266.7	96.0	2.78
P2	324.3	107.0	3.03	532.4	177.0	3.01
P3	518.2	138.5	3.74	698.2	247.0	2.83
P4	426.1	143.0	2.98	524.3	150.0	3.50
P5	301.5	139.0	2.16	265.2	96.0	2.76
P6	513.2	172.0	3.00	483.7	156.0	3.10
P7	731.3	237.5	3.08	861.5	323.5	2.66
P8	370.6	134.5	2.76	376.5	176.0	2.14
P9	585.9	163.0	3.60	526.5	151.0	3.49
P10	320.8	134.5	2.39			
P11	451.9	144.0	3.14			
P12	305.9	120.0	2.55	434.0	150.0	2.89
P13				356.0	112.5	3.16
P14	250.0	77.5	3.22	377.0	101.5	3.71
P30	335.0	139.0	2.41			
P31	275.3	120.0	2.29	292.0	114.5	2.55
P32	456.9	155.0	2.95	458.8	189.0	2.43
P33	384.6	110.0	3.50	368.5	110.0	3.35
P34	366.6	124.5	2.95	377.1	155.5	2.43
P35	435.0	149.0	2.92	373.5	115.0	3.24
P36	314.2	177.0	1.78	206.8	109.5	1.89
P37	579.7	187.0	3.10	846.3	299.5	2.82
P38	378.9	132.0	2.87	541.4	167.5	3.23
P39	191.1	85.5	2.23	268.5	120.0	2.24
P40	197.5	100.0	1.97	172.0	86.0	2.00

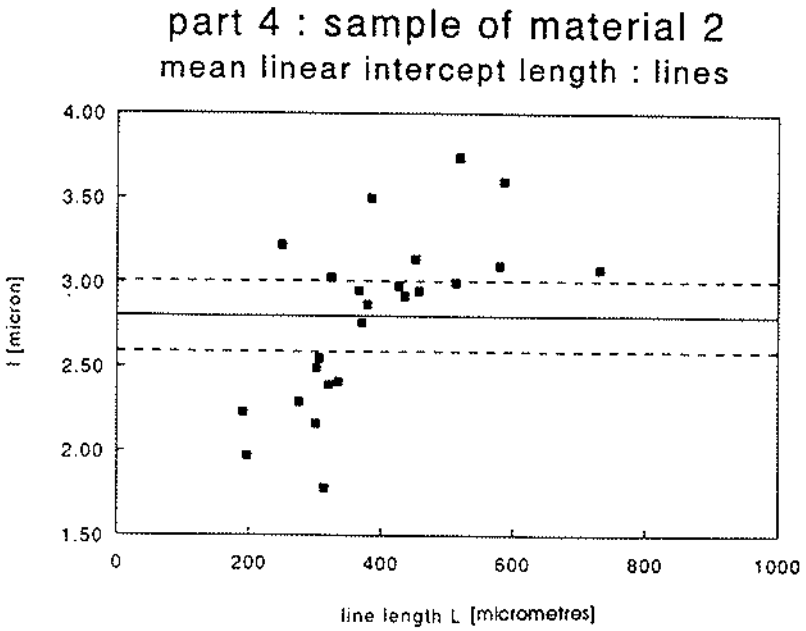

Figure 4.4.1.1 Mean linear intercept length l as a function of the line length L for all participants. ——— = average value for l, — — — = 95% confidence interval

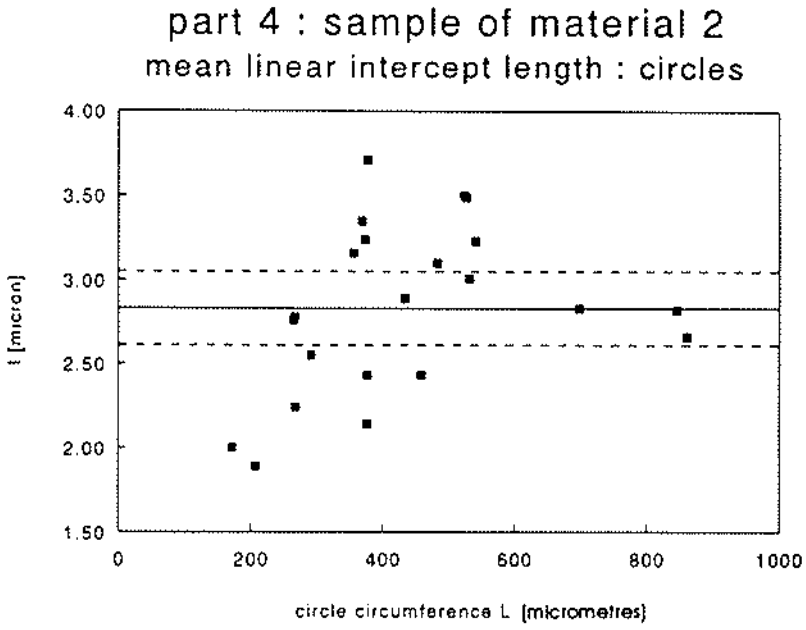

Figure 4.4.1.2 Mean linear intercept length l as a function of the circle circumference L for all participants. ——— = average value for l, — — — = 95% confidence interval.

From Table 4.4.1.1 the following parameter values have been determined:

Lines

Average L for 24 participants	: 388 [μm]
Average n_i for 24 participants	: 138
Average l for 24 participants	: 2.80 [μm]
Standard deviation on average l	: 0.50 [μm]
95% confidence interval for average l (t-distribution with 23 degrees of freedom)	: 2.59..3.01 [μm]

Circles

Average L for 22 participants	: 437 [μm]
Average n_i for 22 participants	: 155
Average l for 22 participants	: 2.83 [μm]
Standard deviation on average l	: 0.50 [μm]
95% confidence interval for average l (t-distribution with 21 degrees of freedom)	: 2.61..3.05 [μm]

From these data is readily found that, again, statistically the line and circle methods do not yield different mean linear intercept lengths (>95% confidence). The scatter between the participants is about 35% which is higher than for Part 1, Part 2 and Part 3.

Some of the factors introducing the scatter found for Part 1, Part 2 and Part 3 have to be re-examined here. The scatter of 10% due to random positioning of lines and circles found in Part 1 will inevitably play a role here too. The result of P4 again may be biased due to the fact that concentric circles were used.

As the micrograph for this Part was prepared by the Participants themselves, as was found in part 2, possibly 15% scatter will be introduced through the preparation procedures as listed in Appendix 4. From these procedures no obvious influence of etching temperature and time could be found. As anticipated by the organisers, material 2 proved to be more difficult for preparation of a quality micrograph, which was confirmed by remarks of some of the participants. Although possible subjective, in view of the organisers the micrographs prepared have a wider range of quality than the ones prepared for material 1 in part 2. Again the range in sizes of the micrographs seems to have no direct influence. Most of the micrographs were taken by SEM with P9, P14, P31, P37 and P38 using an optical microscope. The quality of the micrographs of P9, P14 and P37 in terms of sharpness of grain boundaries was clearly less than those of the other participants. P33 has redrawn the grain boundaries with a thin pencil. The results of P9, P14, P33 and P37 may therefore be more extreme as smaller grains and grain boundaries are more difficult to see. In comparison with the other participants, the micrograph of P3 seems to contain a relatively large number of large grains, whereas for P36 and P40 the opposite holds. This might explain their relatively high and low values, respectively, for the mean linear intercept length. The micrograph of P38 shows some clearly visible grain pluck-out.

Finally as observed in Part 3, 10% scatter may be introduced due to the interpretation of a

micrograph with a more complicated structure.

If these 3 sources of scatter are added the total amounts about 35% which would explain the scatter given above. This can be checked by calculation of the mean results if results for participants P3, P4, P9, P14, P33, P36, P37 and P40 are excluded for the reasons given above. Then, for the line method the value of l is 2.70 micrometres with a standard deviation of 0.33 micrometre, while for the circle method one obtains a mean of 2.73 micrometre with a standard deviation of 0.36 micrometre. These values give a scatter of about 25%, similar to that found in Part 2 and Part 3. The conclusion could then be that, in principle, the same amount of scatter is found if some precaution is taken in evaluation of the results.

4.4.2 Determination of grain size distribution

As was done in Part 3, the grain size distribution was also determined Part 4 using the two grid methods. Again, as a first check on the results, the average intercept size l and the standard deviation s of the grain size distribution returned by each participant, will be discussed. Table 4.4.2.1 and Figures 4.4.2.1 and 4.4.2.2 show the data obtained.

Table 4.4.2.1 Results for grain size distribution measurement.

(N = number of grid intersections, l = mean intercept size [μm], s = standard deviation of intercept sizes [μm])

	Method 1			Method 2		
Participant	N	l	s	N	l	s
P1	217	2.83	2.69			
P2	350	3.65	3.21	422	3.62	3.09
P3	141	3.71	3.42			
P4	268	3.58	2.85			
P5	404	2.86	2.91	484	2.90	3.00
P7	548	3.06	2.93	1178	2.76	2.61
P8	382	3.74	2.97			
P9	318	3.40	3.30	369	3.50	3.30
P10	241	2.10	2.00	432	2.00	1.90
P11	568	2.90	2.65			
P12	311	2.93	2.87			
P14	316	3.43	3.09	431	3.42	2.97
P30	210	2.76	2.48			
P32	319	2.64	2.47			
P33	133	3.50	3.40	260	3.30	3.30
P36	267	2.14	2.00	411	2.02	1.89
P38	182	3.46	3.38	236	3.53	3.34
P39	154	2.30	1.70			
P40	207	1.76	1.41			

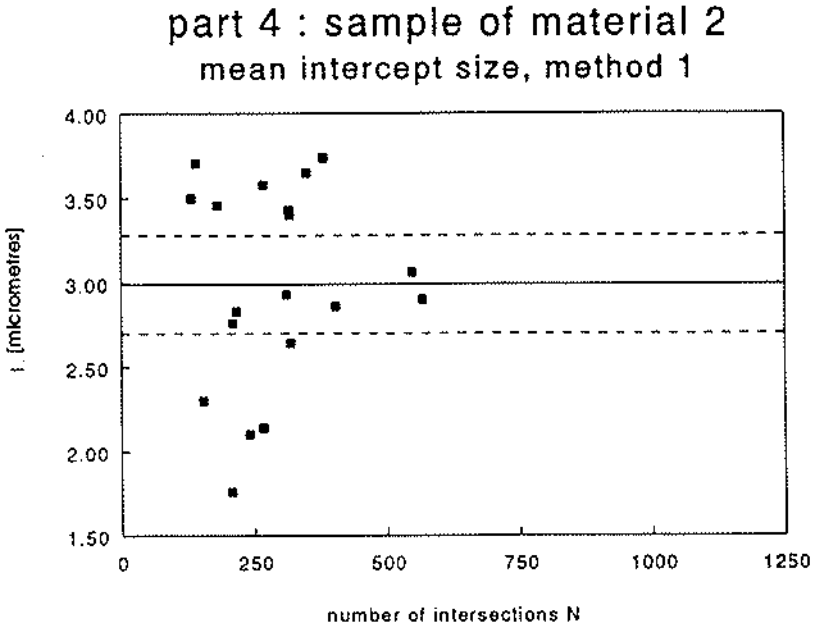

Figure 4.4.2.1 Mean intercept size l as a function of the number of grid intersections N for all participants for Method 1. ——— = average value for l, — — — = 95% confidence interval.

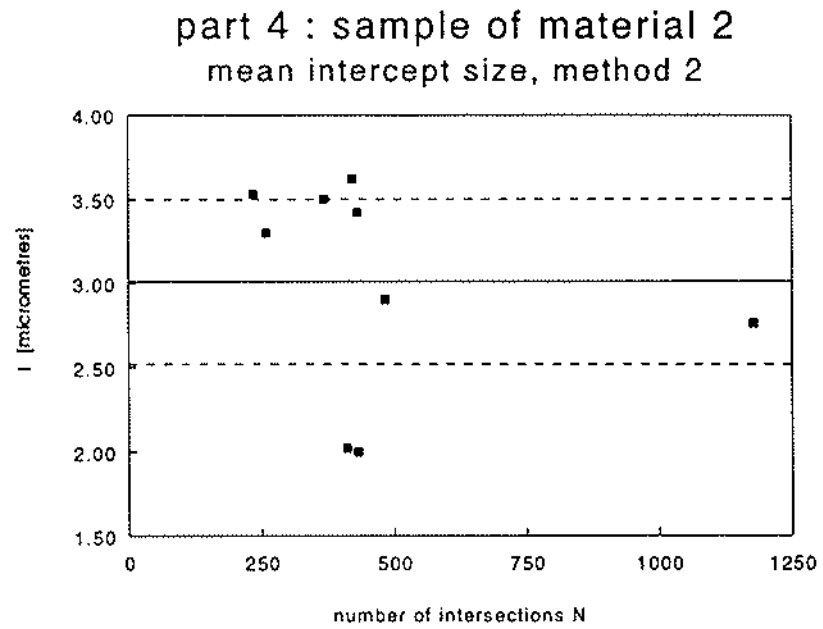

Figure 4.4.2.2 Mean intercept size l as a function of the number of grid intersections N for all participants for Method 2. ——— = average value for l, — — — = 95% confidence interval.

From Table 4.4.2.1 the following parameter values have been determined:

Method 1

Average N for 19 participants	: 291
Average l for 19 participants	: 2.99 [μm]
Sample standard deviation	: 0.60 [μm]
95% confidence interval for average l (t-distribution with 18 degrees of freedom)	: 2.70..3.28 [μm]

Method 2

Average N for 9 participants	: 469
Average l for 9 participants	: 3.01 [μm]
Sample standard deviation	: 0.64 [μm]
95% confidence interval for average l (t-distribution with 8 degrees of freedom)	: 2.52..3.50 [μm]

Using the 95% confidence interval for the mean intercept length determined on the same micrograph with the line method (Section 4.4.1) of 2.59..3.01, it is readily seen that the values for the average l given above are within this confidence interval. This again indicates that the mean value of the grain size distributions determined agrees well with the one obtained with the line method. Again, possible differences between Method 1 and Method 2 are difficult to determine. Also, the scatter in l is of the same order of magnitude (40%) as for the line and circles methods.

As was done for Part 3, the grain sizes measured were plotted on a lognormal scale basically yielding the same type of figures as discussed in Section 4.3. Again the graphs were fitted to obtain the 50% probability grain size x_{50} and the standard deviation σ_{lnx} of the lognormal distribution. The results are as listed in Table 4.4.2.2, while the results for x_{50} are shown in Figures 4.4.2.3 and 4.4.2.4.

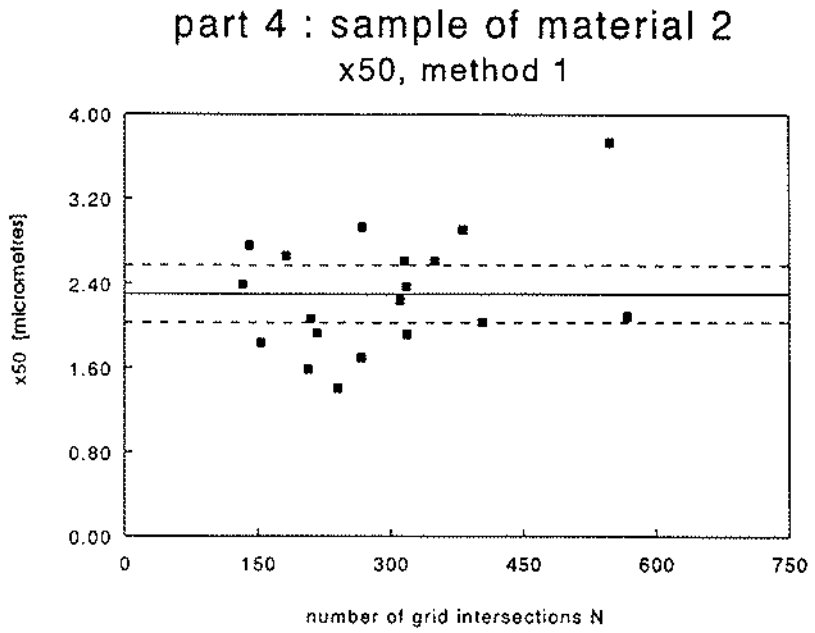

Figure 4.4.2.3 Computed values for 50% probability grain size x_{50} for Method 1. ——— = average value for x_{50}, — — — = 95% confidence interval.

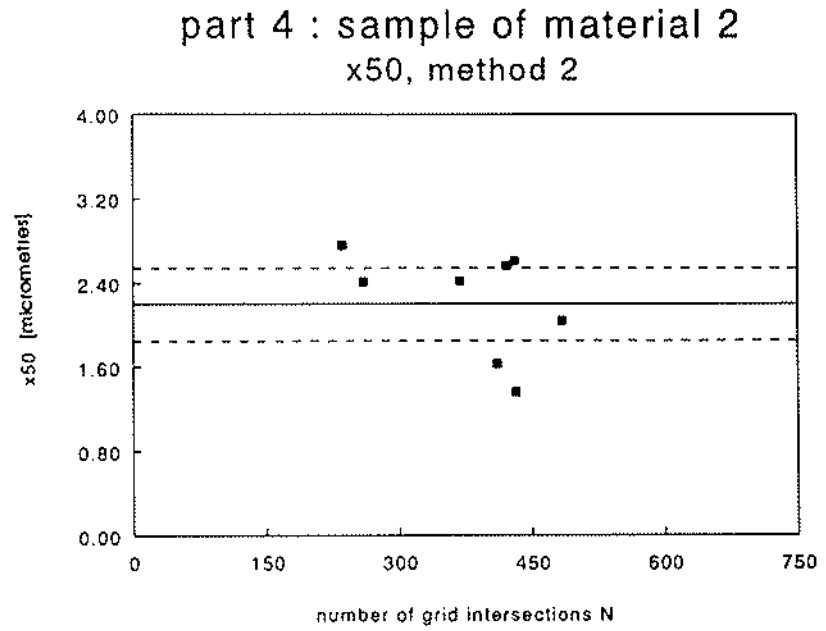

Figure 4.4.2.4 Computed values for 50% probability grain size x_{50} for Method 2. ——— = average value for x_{50}, — — — = 95% confidence interval.

Table 4.4.2.2 Computed values for 50% probability grain size x_{50} [μm] and standard deviation σ_{lnx}.

Participant	Method 1		Method 2	
	x_{50}	σ_{lnx}	x_{50}	σ_{lnx}
P1	1.93	0.86		
P2	2.61	0.80	2.56	0.80
P3	2.76	0.87		
P4	2.93	0.70		
P5	2.03	0.88	2.04	0.87
P7	3.73	0.88	2.01	0.82
P8	2.90	0.75		
P9	2.37	0.81	2.42	0.81
P10	1.41	0.92	1.36	0.89
P11	2.09	0.81		
P12	2.24	0.81		
P14	2.61	0.77	2.61	0.75
P30	2.07	0.75		
P32	1.92	0.82		
P33	2.39	0.88	2.41	0.85
P36	1.70	0.77	1.63	0.75
P38	2.66	0.82	2.76	0.79
P39	1.84	0.65		
P40	1.59	0.65		

From Table 4.4.2.2 the following mean values and standard deviations were obtained:

Method 1

Mean value for x_{50} : 2.30 [μm]
Sample standard deviation : 0.55 [μm]
95% confidence interval for average x_{50} : 2.03 .. 2.57 [μm]
(t-distribution with 18 degrees of freedom)

Method 2

Mean value for x_{50} : 2.20 [μm]
Sample standard deviation : 0.45 [μm]
95% confidence interval for average x_{50} : 1.85 .. 2.55 [μm]
(t-distribution with 8 degrees of freedom)

The scatter in the values for x_{50} is about 40 to 50% which is somewhat higher than the scatter in the values for the mean linear intercept length and the values for the mean grain size. Part of this will be due, of course, to the use of a fitted parameter. From the discussion on the results for the mean linear intercept length, it will also be clear that some of the results for the grain size distribution measurement will be more extreme for

more or less understandable reasons (P9, P14 and P33: quality of micrograph; P3, P36 and P40: many large or small grains). In addition it has to be mentioned that P8 used a relatively small grid of 5*5 mm on a micrograph of 115*90 mm whereas most of the other participants used a grid of 10*10 or 20*20 mm. The small grid size may lead to a larger mean grain size as discussed for Part 3, where the same phenomenon was observed. The results of these participants do not explain the large scatter in x_{50}, however, as omitting them has only marginal influence. Omitting these results in the calculation of the average value for l from Table 4.4.2.1 gives an average value for l of 2.92 and 2.96 μm for Method 1 and Method 2 respectively, which is 10% higher than for the line and circle methods, and reduces the scatter in the average value for l for Method 1 from 40 to 30%, which is still somewhat higher than the scatter found in the Parts discussed sofar.

As was done for Part 3 the grain sizes measured were combined to a single distribution as shown in Figures 4.4.2.5 and 4.4.2.6.

As can be seen from these Figures, Method 1 and Method 2 do not yield a largely different result.

In summary, one could conclude that the values for the average grain size are consistent with the values obtained by the line and circle methods if some precautions are taken. This especially applies to the micrograph quality and whether the micrograph has been taken from a comparable portion of the specimen such that a comparable portion of the actual grain size distribution is taken into consideration. The grain size distribution itself is subjected to more scatter (40-50%) which will be due to a combination of scatter introduced due to micrograph preparation and scatter due to interpretation of the micrograph. As the scatter for these analyses in Part 3 amounted 30%, it seems justifiable to assume that 10 to 20% additional scatter is obtained by the influence of micrograph preparation.

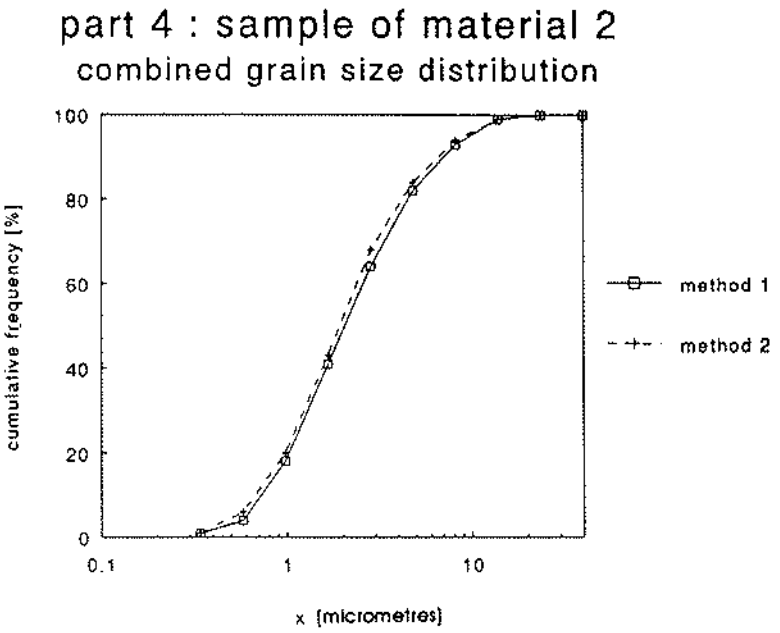

Figure 4.4.2.5 Combined cumulative frequency distribution for grain size determined with Method 1 and Method 2.

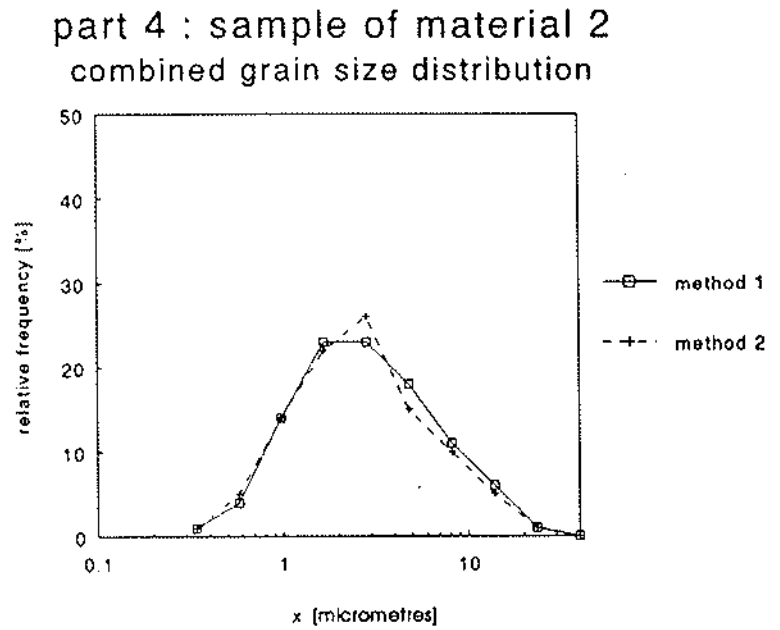

Figure 4.4.2.6 Combined relative frequency distribution for grain size determined with Method 1 and Method 2.

4.5 Results from image analysis systems

Only few of the participants returned results obtained with an image analysis system, namely P3, P8 and P34. P3 and P34 obtained grain sizes as the equivalent grain diameter (the diameter of the circle having the same surface as the grain), while P8 obtained grain sizes as the length of intersections of vertical lines with the grains. As these methods will in general not produce the same results as the manual methods discussed sofar, a comparison of the results is difficult. Therefore the results obtained with automatic image analysis systems will be given and discussed as far as possible. Of course a statistical analysis can not be undertaken because of the limited amount of data.

Part 1 computer drawn micrograph

P3 : d_{eq} = 9.1 mm (233 grains)
P8 : l = 6.27 mm
P34 : d_{eq} = 9.4 mm

Part 2 sample of material 1

P3 : d_{eq} = 3.3 μm with a standard deviation of 1.7 μm
P8 : l = 1.1 μm
P34 : d_{eq} = 3.4 μm

Part 3 micrograph of material 2

P3 : d_{eq} = 5.2 mm with a standard deviation of 4.4 mm
P34 : d_{eq} = 4.5 mm with a standard deviation of 4.5 mm

Part 4 sample of material 2

P3 : d_{eq} = 3.1 μm
P8 : l = 0.6 μm
P34 : d_{eq} = 2.8 μm with a standard deviation of 2.2 μm

In general, the values do not compare with the values obtained with the line and circle methods. The method used by P8 is more or less like the manual methods but shows lower values. The results of P3 and P34 are in general higher. For Part 1 this can be explained. The micrograph has a surface of 159*159 mm^2 and contains 350 grains, yielding an average grain size of 72.2 mm^2. This would lead to an equivalent diameter of 9.6 mm, which is much higher than the value for the mean linear intercept length of about 7 mm. The results of P3 and P34 are comparable to this value. Also for the other tasks, the results of P3 and P34 are well comparable.

In view of the limited amount of data, no conclusions can be drawn on these results. A more elaborate Round Robin exercise on the use of image analysis systems would therefore be necessary to assess whether these methods can be included in future standards. Such a standardization may turn out necessary with the progress being made in

the field of image processing and the introduction in increasing numbers of these systems for routine use.

5. Conclusions and recommendations

The results of a Round Robin exercise for grain size measurement of advanced technical ceramics have been discussed and analysed. Prime targets of this Round Robin were to validate a draft CEN standard for mean linear intercept length measurement and to test methods for manual determination of porosity and grain size distribution. Twenty-five companies, institutes and universities from Europe, Japan and the USA participated.

Part 1 of the Round Robin used an ideal computer generated microstructure. Analysis of the results showed that the line and circle methods for mean linear intercept length measurement yield the same value with a scatter of about 10%. Premises are that sufficient intersections are counted (> 100) and that random positioning of lines and circles is guaranteed. The scatter in Part 1 may be seen as the minimum attainable scatter in these types of measurements and is due to the influence of random positioning of a finite number of lines and circles.

Part 2 of this exercise used a sample of material 1 with a "nice" microstructure (equiaxed grains of relatively uniform size with clearly marked grain boundaries). The results show a scatter of about 25%. This 15% increase in scatter with respect to Part 1 may be due to the influence of micrograph preparation and selection of analysis area only. It is likely that undesired grain growth plays a disturbing role in some of the results and also the large variety in polishing and etching procedures applied may have increased the scatter.

Part 3 of the Round Robin dealt with a micrograph of material 2 with a more complicated microstructure which was sent to all participants. Porosity was measured with a grid adopted as a comprise between being fine enough for counting sufficient pores and being coarse enough to avoid eye-strain. The scatter in the results of about 60% is relatively large but can be due to the fact that the porosity is a small number (2.4% as an average value). Major sources of scatter will be the influence of random positioning of the grid and the interpretation of what is a pore. The mean linear intercept length was measured using the line and circle methods, and showed a scatter of 20% after some results were excluded because less than the required 100 intersections were counted. The 10% increase in scatter with respect to Part 1 can be attributed to interpretation of a more complicated microstructure having a wider grain size distribution. Also, grain size distributions were measured using a grid method. The mean grain size corresponds well to the mean linear intercept length determined with the line and circle methods. Again the scatter in the results is about 30% after some results were excluded because a much finer or coarser grid was used. No difference between grid Method 1 (forbidden lines) and Method 2 (counting all grains) could be found.

Part 4 of the Round Robin used a micrograph of material 2 prepared by each participant. The mean linear intercept length was measured with the line and circle methods showing a scatter of about 25% after some results were excluded because of the quality of the micrograph or because a portion of the material with relatively many small or large grains

was sampled. The 15% increase in scatter with respect to Part 1 can be assigned to the influence of micrograph preparation and interpretation of a more complicated microstructure, and is likely to be a combination of additional scatter found in Part 2 and Part 3 (15 and 10% respectively). Again, grain size distributions were measured showing a scatter of about 30% with no clear difference between Method 1 and Method 2.

One of the targets of this Round Robin was to validate the draft CEN standard for grain size measurement using linear intercept methods. From the results given it can be concluded that in principle these methods yield identical results if some precaution with respect to the number of grain intersections and the randomness of lines and circles is taken. Using about 100 grain intersections gives a scatter of about 10% which is considered as a reasonable number. Additional scatter can be introduced however depending on micrograph preparation and the degree of complexity of the microstructure. From the results given it may be concluded that a scatter of 20 to 25% is likely to be found. Whether this is an acceptable number will depend on the actual application. Part of the scatter can be reduced by taking into account more grain intersections although it can be expected that in case 1000 intersections are used this will only decrease the scatter by about 5% if the randomness found in Part 1 is inversely proportional to $\sqrt{n_i}$. Therefore, in any practical situation, the influence of micrograph preparation and micrograph interpretation will control reliability of the results. Experience and knowledge then become dominating factors which cannot be standardized.

The second target of this Round Robin was to test methods for measurement of porosity and grain size distribution. The grid method for porosity measurement reveals the basic problem of how to measure a small number accurately. The grid used was considered fine enough but still substantial scatter was found. Therefore a coarser grid might have been just as appropriate. To obtain a reasonable estimate for the porosity this method may be sufficient however, and certainly so if the level of scatter can be reduced by counting on different parts of the material with different positions of the grid.

The grid methods for grain size distribution measurement seem to yield relatively consistent results. Expected differences between Method 1 and Method 2 are not found, so they are likely to be of less influence than a factor as micrograph preparation e.g. The amount of scatter in the results is in general comparable to the scatter found for the measurement of mean linear intercept length. As such no objection seems to exist to the methods proposed.

From the results discussed it can be concluded that micrograph preparation and interpretation, which are not necessarily independent factors, are of major importance for the amount of scatter. The large scatter for Part 3 possibly is due to the fact that the micrograph provided is not of an optimal quality, which was done on purpose by the organisers. But the even larger scatter in Part 4 again shows the influence of micrograph preparation. From the assembled collection of micrographs returned by the participants striking differences in magnification, contrast amount of reflections on SEM images etc are found. It therefore is suggested that some typical "good" and "bad" images are included in standards as guidelines for users. Also, it is believed that the influence of the etching procedure should be more specifically addressed as it is likely to have a major

influence on the micrograph quality. This also applies to details of the set-up of optical and SEM microscopes.

The methods discussed in this report in principle also can be used for determination of phase content and grain size distribution of multiphase materials. Whether this yields comparable results as discussed in this report is uncertain. It would be well worthwhile to make this a topic for a future VAMAS or joint CEN/VAMAS Round Robin exercise to provide information useful for formulation of standards on these important topics.

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Appendix 1 : Draft CEN standard for grain size measurement

EUROPEAN STANDARD

METHODS OF TEST FOR ADVANCED MONOLITHIC TECHNICAL CERAMICS

ENV 623: GENERAL AND TEXTURAL PROPERTIES

Part 3: Determination of Grain Size

CONTENTS:

1. SCOPE

This Part of ENV 623 describes procedures for polishing and etching test-pieces of advanced technical ceramics, followed by the preparation of micrographs of the microstructure and methods of making measurements for the determination of grain size.

NOTE 1: This standard applies to single-phase ceramics, and to ceramics with a principal crystalline phase and a glassy grain-boundary phase of less than about 10% by volume. The measurements described do not apply to ceramics with more than about 10% by volume of pores or of continuous glassy phase, nor to ceramics with more than one crystalline phase. Such materials require alternative methods which allow the pores or phases to be distinguished and counted separately, and this is beyond the scope of this standard.

NOTE 2: This standard does not cover automatic image analysis methods of determining grain size and other parameters. By agreement between parties, such methods may be used as an alternative to this Standard, but the report must state explicitly that such techniques have been used, and give the basis of the method, which may not be the same as in this Standard (see 7.).

2. NORMATIVE REFERENCES

This European Standard incorporates, by dated or undated reference, provisions from other publications. These normative references are cited at the appropriate places in the text and the publications listed hereafter. For dated references, subsequent amendments to or revisions of any of these publications apply to this European Standard only when incorporated in it by amendment or revision. For undated references the latest edition of the publication referred to applies.

EN ---- Sampling of advanced monolithic technical ceramics.

3. DEFINITIONS

3.1 Grain size

The grain size is the size of the distinct crystallites in a material, and for the purposes of this method of test, is that of the primary or major phase.

NOTE: In materials which contain more than one phase, it may be necessary to characterise the different phases. The phases may be continuous or as isolated grains. In this method, only the primary phase is characterised, and minor phases are ignored.

3.2 Mean linear intercept grain size (g_{mli})

The mean linear intercept size is the average value of the distance between grain boundaries as shown by randomly positioned lines drawn across a micrograph or other image of the microstructure.

4 APPARATUS REQUIRED

4.1 Sectioning equipment

A suitable diamond-bladed cut-off saw is required to prepare the initial section for investigation.

4.2 Mounting equipment

Suitable metallurgical mounting equipment and media are recommended for providing firm gripping of the test-pieces for polishing.

4.3 Grinding and polishing equipment

Suitable grinding and polishing equipment employing diamond abrasive media is required.

NOTE 1: Annex 1 recommends techniques and abrasives.

4.4 Microscope

An optical or scanning electron microscope with photomicrographic facilities. For materials with a mean linear intercept grain size of less than about 4 μm a scanning electron microscope is recommended in order to obtain sufficient magnification of the photomicrograph. A reference graticule is required for determination of magnification in an optical microscope, and a reference square grid or latex spheres are required for calibration of magnification in a scanning electron microscope. In all cases, the calibration of dimensions of the references shall be traceable to national or international standards of length measurement.

NOTE 2: ASTM E766 describes a practice suitable for the scanning electron microscope.

5. TEST-PIECE PREPARATION

5.1 Sampling

The test-pieces shall be sampled in accordance with the guidelines given in ENV ----, and subject to agreement between parties.

NOTE 1: Depending on the objectives of performing the measurement, it is desirable to maintain full knowledge of the positions within components or test-pieces from which sections are prepared.

5.2 Cutting

The required section of the test-piece shall be cut using a diamond saw, preferably a

metal-bonded saw with a diamond grit size not greater than D151[1] (approximately 125-150 μm), employing adequate coolant.

NOTE 2: For routine inspection of materials, a small area of not more than 10 mm side is normally adequate as the section to be polished. Large areas generally take longer to polish.

5.3 Mounting

Mount the test-piece using an appropriate proprietary mounting medium. If the material has significant open posity (>30%) it is advisable to vacuum impregnate the test-piece with liquid mounting resin before encapsulating as this will provide some support during polishing.

NOTE 3: It is not essential to encapsulate the test-piece. For example, it could be affixed to a metal holder. However, encapsulation in a plastic-based medium allows easy gripping and handling, especially of small irregularly shaped test-pieces and of porous, friable materials.

5.4 Grinding and polishing

Grind and polish the surface of the test-piece. Care should be taken to ensure that grinding produces a planar surface with a minimum of damage. Successively smaller grit sizes shall be employed, at each stage removing the damage from the previous stage until there is no change in appearance when examined by an optical microscope at high magnification. The final surface shall be free from optically visible scratches or other damage introduced by polishing.

NOTE 4: Care should be taken in choosing the sequence of grits and lap types. It is impossible within the scope of this Standard to make specific recommendations for all types of material. The general principle to be adopted is the minimisation of subsurface damage, and its removal by progressively finer grits whilst retaining a flat surface. Some guidelines on grinding and polishing are given in Annex 1.

5.4 Etching

When a good quality surface has been achieved, the test-piece shall be etched if necessary to reveal grain boundaries. Any suitable technique shall be used, subject to agreement between parties.

NOTE 5: Some general guidelines recommending etching procedures for various commonly available advanced technical ceramics are given in Annex 2.

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[1]Federation of European Abrasives Manufacturers (FEPA) designation

6. PHOTOMICROGRAPHY

6.1 General aspects

If it is suspected that the average grain size of the test material is less than about 4 μm, it will be necessary to prepare the test-piece for scanning electron microscopy. Otherwise, optical microscopy will generally be adequate.

6.2 Optical microscopy

Set up Kohler illumination in the microscope.

> NOTE 1: A procedure for doing this will normally be found in the documentation for the microscope. Alternatively, a proedure may be found in ASTM F728.

Examine the test-piece at a magnification sufficient to resolve the individual grains clearly. If the contrast obtained is insufficient, e.g. in white or translucent materials, apply a suitable metallic coating by evaporation or sputtering. Prepare micrographs of at least three different areas of the test-piece surface. As a guideline, there should be no more than 20 grains along any 75 mm line drawn on the micrograph so produced. If there are more grains than this, increase the magnification and prepare fresh micrographs. Unless already undertaken, prepare a micrograph of a graticule at the same magnification to provide a calibration of magnification. Micrographs should be of at least 100 x 75 mm size.

6.3 Scanning electron microscopy

If the test-piece is not electrically conducting, mount the test-piece on the test-piece holder of the microscope and apply a thin evaporated or sputtered conductive coating. Insert the test-piece into the microscope, ensuring that the surface to be characterised is normal to the electron beam to within 5°.

> NOTE 2: This ensures that the image does not suffer from excessive distortion due to angle of viewing.

Prepare micrographs at a suitable magnification (see 6.2) from at least three different areas of the test-piece. For calibration of the lateral and vertical magnification of the micrographs prepare similar images of a graticule or grid, or of calibrated spheres at the same working height of the microscope stage.

> NOTE 3: The photographic screen in the microscope may not have constant magnification at all points. A square grid makes a suitable reference for ascertaining the degree of distortion in the screen, since it is easy to detect distortions of the grid. If the image distortion is uniform across the field of view, i.e. lateral and vertical magnifications appear to be constant but different, it is possible to make corrections when measuring the micrographs (see Note 2 in 7.). For the purposes of this standard, the actual magnification should not vary by more than 5% over the area of the screen.

7. MEASUREMENT OF MICROGRAPHS

Draw at least five thin straight lines of length exceeding 75 mm and of random position and orientation across each micrograph, giving a total line length of not less than 375 mm intersecting at least 100 grains. Measure each line length to the nearest millimetre and calculate the total line length (l_t). Count the number (n_i) of intersections of the lines with grain boundaries. If the line intersects the junction of three grains, count this as 1.5 intersections. If the line intersects a large pore, count this as one intersection. Measure the total length of line that crosses large pores (l_p). If the line runs along a grain boundary, count this as one intersection.

Alternatively, on each micrograph draw at least two circles of diameter not less than 10 times the expected mean grain size using a pair of compasses and randomly positioning the centres. Measure the diameters of the circles (d) to the nearest 0.5 mm, and calculate the sum of their circumferences (l_t). Count the number (n_i) of intersections of each circle with the grain boundaries. If the intersection coincides with the junction of three grains, count this as 1.5 intersections. If the line intersects a large pore, count this as intersection. Measure the approximate circle circumference length that crosses large pores (l_p).

NOTE 1: For the purposes of this standard, a large pore is one which resides at grain boundaries. Small pores entrained within grains should be ignored.

Check the calibration of the micrographs in the following manner. For optical microscopy, measure the size of the spacing of the calibrated graticule as shown by a micrograph and calculate the apparent magnification. For scanning electron micrographs employing a graticule or grid, use the same procedure to calculate the magnification horizontally and vertically. If calibration spheres have been used, measure the horizontal and vertical dimensions of at least six spheres and calculate the respective mean values. If the vertical and horizontal magnifications calculated are different by more than 5% or individually vary by more than 5% across the screen, the distortion of the image is not acceptable for the purposes of this Standard.

NOTE 2: If image distortion in the scanning electron microscope is uniform across the field of view, i.e. horizontal and vertical magnifications are constant but different, the effective magnification of each drawn line can be calculated by noting its angle relative to the horizontal on the micrographs and applying an angular correction to the magnification. This procedure may only be adopted by agreement between parties.

NOTE 3: If it is desired to apply an automatic image analyser to the measurement of micrographs or direct images, in order that the results are comparable with the manual method described in this standard the following points must be noted.

(a) The image must be scanned in at least five random directions, which may be achieved either through software design or by rotating the image to random positions. Scanning in only one direction on the test-piece is not acceptable since it does not allow for anisotropy.

(b) Care must be taken that the contrast change at a grain boundary is sufficient for the detection system.

(c) The calculation routine incorporated in the software must operate in the same way as this manual method in order that large pores are discounted.

Failure to observe these points will result in results which may be substantially at variance with the manual method.

8. CALCULATION OF RESULTS

For both line and circle methods, calculate the mean linear intercept distance, g_{mli}, in micrometres for each micrograph using the formula:

$$g_{mli} = \frac{(l_t - l_p).10^3}{n_i.m}$$

where: l_t is the total line length in millimetres; in the case of circles, the total circumference of the circles;
l_p is the total line length that crosses large pores;
n_i is the counted number of intersections on each micrograph;
m is the calibrated magnification of the micrograph.

9. INTERFERENCES AND UNCERTAINTIES

The nature of microstructure of the test-piece can affect the result determined by this test, especially in cases where there is a wide distribution of grain sizes (e.g. a bimodal distribution), or where it is difficult to find an adequate etching method to reveal grain boundaries. The method also assumes that the amount of continuous secondary phase is small compared with the major crystalline phase(s). As the widths of the layers of such secondary phase between grains of the primary phase increase, there will be increasing overestimate of true mean grain size. The method also assumes that the total porosity level is negligible.

The principal causes of uncertainty in this method are considered to be the random errors of selecting areas of the test-piece from which to prepare micrographs and the positions on the micrograph in which to draw lines or circles. The former depends on the homogeneity of the microstructure within the test-piece, and the latter on any subjective element in selecting line or circle positions.

Uncertainties arising from magnification and counting are considered to be relatively unimportant provided that thes procedure described in this standard are followed.

10. REPORT

The report of the test shall contain the following:

(a) the name of the testing laboratory;

(b) a unique identification of the report;

(c) the name and address of the client;

(d) details of the test-piece, including material type, manufacturing code, batch number, etc.;

(e) the date of receipt of the test item(s) and of the test;

(f) a reference to this Standard, i.e. ENV ---- part 3;

(g) the observation technique employed (optical or scanning electron microscope);

(h) the technique employed for calibration, and the resulting magnification;

(i) a summary of the procedure for sampling, cutting, grinding, polishing and etching the test-piece;

(j) copies of the micrographs used for the measurement;

(k) whether lines or circles were used for the analysis, and the total line length employed for the measurements, expressed in millimetres;

(l) the number of intercepts for each of the five lines or two circles on each of the three micrographs;

(m) the total number of intercepts;

(n) the calculated mean linear intercept size for each of the micrographs, expressed in micrometres;

(o) any remarks on the general appearance of the microstructure, whether isotropic or anisotropic, the presence of secondary phases, whether the grain size is obviously bimodal, or the grain shape is anisotropic;

(p) signature of person responsible for the test and authorising issue of report

NOTE: For routine presentation of results it is useful if a standardised format is adopted. A recommended scheme is presented in Annex 3.

References

ASTM E766 Practice for calibrating the magnification of a scanning electron microscope using NIST-SRM-484.

ASTM F728 Preparation of an optical micrcroscope for dimensional measurements, standard practice for.

Annex 1 - Grinding and polishing procedures

Preparation of polished sections of ceramics requires different procedures from those conventionally employed for metallic materials, which typically commence with a coarse grinding stage using fixed grit silicon carbide papers of grit sizes of 30 μm or greater. For ceramic materials, this type of procedure can produce considerable amounts of sub-surface damage in the form of extended microcracks which can then influence the microstructural appearance obtained, unless precautions are taken to minimise such damage and to remove all traces of it in subsequent grinding steps. Unless care is taken, the final surface may contain damage which manifests itself as microcracks and grain tear-out, the presence of which can influence the results of any microstructural characterisation measurement. Thus, selection of appropriate polishing procedures, including the sequence of grit sizes, the times of abrasion, and the applied pressure are all important. Optimum conditions vary considerably depending on the type of material being prepared. Guidelines on how to choose a grinding method may be found in Hubner and Hausner [A1.1].

As an example, a series metal-bonded diamond grinding discs give high material removal rates for initial flattening. However, grit sizes greater than 30 μm may introduce damage, especially in materials of poor toughness, and smaller grit sizes used for longer periods of time may produce a better result. Loose diamond abrasives remove material more slowly than grit of the same size fixed in a discs, and may cause more damage. Subsequent grinding steps may need to be of longer duration. The use of a shock-absorbing system, such as a soft metal lap (e.g. tin) into which loose grit becomes lodged, or a metal-plastic composite lap with fixed diamond grit, gives a good balance between speed of abrasion and surface damage.

The grinding of silicon carbide ceramics can cause special difficulties. Klimek [A1.2] recommends that the diamond abrasive used should not be larger than 6 μm, since a larger size of abrasive tends to shatter large SiC grains rather than to produce cutting.

After a planar surface is achieved with the initial grinding stage, a sequence of finer grit sizes may be employed to remove grinding damage from previous steps. The precise sequence of stages chosen will depend on equipment available, and may have to be optimised for each type of material. The general principle should be that each step should be of sufficient duration to remove evidence of damage from the previous stage. The final polishing stage should not be undertaken until a good quality finish is obtained. The use of napped cloths for polishing is not recommended because on many types of ceramic it can cause pluck-out of grains (especially with high-alumina ceramics) or loss of flatness of surface. Polishing procedures have been described by Clinton [A1.3]. A series of articles on microstructural preparation of ceramics with polishing details is given in Reference [A1.4].

The following five-stage procedure is recommended as a starting point for fine-grained ceramics, and gives surfaces of sufficient quality for examination at high magnification in the scanning electron microscope:

1. 30 μm diamond on a hard composite lap;
2. 6 μm diamond on a softer composite lap;
3. 1 μm diamond on a hard napless cloth (or a tin lap);

4. 0.25 μm diamond on a hard napless cloth;
5. colloidal silica in alkaline solution on a hard napless cloth.

NOTE: The last step is intended to remove scratches from the polished surface, which it does very successfully. However, there is a risk of pores becoming filled with polishing debris which is impossible to remove, and this step should not be used if evaluation of porosity content is required. Such pick-up should not influence grain size measurement. It is recommended that the lap is kept wet at all times, and that polishing is continued with water for a short while at the end to prevent the build-up of deposits on the surface.

Before moving from one stage to the next, the test-piece should be carefully cleaned of abrasive grit using an ultrasonic bath and a suitable liquid cleaning agent, and should be examined in an optical microscope to ensure the surface is uniform and that damage from the previous stage is minimised.

References

A1.1 Hubner , G., Hausner, H., Material-orientated preparation of sintered ceramic bodies, **Prakt. Metallogr.**, 1983, **20**, 289-296.

A1.2 Klimek, E.J., Microstructure of silicon carbide materials. **Microstructural Science, Volume 16**, ASM International, Monterey, USA, 1987, 295-304.

A1.3 Clinton, D.J., **A guide to polishing and etching of technical and engineering ceramics**, Institute of Ceramics, Stoke-on-Trent, Staffs, U.K., 1987.

A1.4 Carle, V., et al., Ceramography of high-performance ceramics - description of materials, preparation, etching techniques and description of microstructures: - Part II: Silicon carbide. **Prakt. Metallog.**, 1991, **28**, 420-34. Part III - Zirconium dioxide (ZrO_2) (by Schäfer, U., et al.), *ibid*, 1991, **28**, 468-83. Part IV - Aluminium nitride (AlN) (by Predel, F., et al.), *ibid*, 1991, **28**, 542-52.

Annex 2 - Etching procedures

With many ceramic materials it is necessary to reveal the positions of grain boundaries for the purpose of this test. A variety of techniques are available for doing this, but the choice and the severity of the process may depend on the precise nature of the material and the technique used to observe the microstructure. Some experimentation is often needed to set appropriate conditions for unfamiliar materials. Over-etching is to be avoided, since it can modify the appearance of the microstructure. It is recommended that the optimum etching conditions are determined in a step-wise fashion to ensure that over-etching does not occur. It may be necessary use more severe etching for SEM images than for optical images in order to produce adequate contrast at grain boundaries.

Bibliographic lists of etching methods have been given by Clinton [A2.1] and Petzow [A2.2], and further information is given in references [A2.3] to [A2.5]. Table 1 shows some examples.

Ceramic	Method	Conditions
Alumina (>99.5%)	Thermal	1500 °C, 2h
Alumina (lower purity)	Chemical or thermal	10 % HF, 20s; 1450 °C, 1h
Zirconia-toughened alumina	Thermal	1500 °C, 15 min
Yttria-TZP	Thermal	1300 °C, 2 h to 1420 °C, 15 min
Ce-TZP	Thermal	1450 °C, 5 min
Sialons and sintered silicon nitrides	Plasma etch [A2.6, A2.7]	Fluorine ions
Hot-pressed silicon nitride	Chemical	NaOH, 400-450 °C, 1-10 min
Aluminium nitride	Relief polished	Colloidal silica, alkaline solution
Sintered silicon carbide	Chemical	Modified Murakami's reagent, e.g. 3 g KOH, 30 g $K_3Fe(CN)_6$, 60 ml H_2O, boil 2-20 min

Thermal etching used for oxide ceramics can give good clear delineation of grain boundaries, but there is a risk of modifying the microstructure of the product in the process. The maximum temperature for this process should be at least 150 K below the original firing temperature of the ceramic to minimise the risks. In addition, the presence of glassy secondary phases can cause problems of contamination of the grain surfaces as it is usually mobile at the required thermal etching temperatures.

Chemical methods, particularly those involving melts, can be difficult to control and reproduce. Ensure that the test-piece is clean and free from grease before using aqueous etchants. If a test-piece is over-etched, smaller grains may disappear. Ceramics with continuous secondary phases are generally more easily etched than those without, but caution is required if the primary phase is also continuous, e.g. in reaction-bonded silicon carbide, or in some high-alumina ceramics. The true grain boundaries may not all be revealed.

References

A2.1 Clinton, D.J., **A guide to polishing and etching of technical and engineering ceramics**, Institute of Ceramics, Stoke-on-Trent, Staffs, UK, 1987.

A2.2 Petzow, G., **Metallographic etching**, American Society for Metals, Ohio, USA, 1979.

A2.3 Elssner, G., et al., **Methoden zur Anschliffspräparation keramische Werkstoffe**, Deutsche Keramische Gesellschaft, Bad Honnef, 1985.

A2.4 Lay, L.A., **Corrosion resistance of technical ceramics**, HMSO, London, 1984.

A2.5 Carle, V., Ceramography of high-performance ceramics - description of materials, preparation, etching techniques and description of microstructures. **Prakt. Metallog.**, 1991, **28**, 359-77.

A2.6 Chatfield, C. and Norstrom, H., Plasma etching of sialon. **J. Amer. Ceram. Soc.**, 1983 **64**(9), C-168.

A2.7 Täffner, U., Hoffman, M.J., Krämer, M., Comparison of different physical/chemical methods of etching for silicon nitride ceramics. **Prakt. Metallog.** 1990, **27**, 385-90.

Annex 3 - Results sheet

Testing establishment		
Report reference number		
Client name and address		
Date of receipt of test-piece		
Date of test		
Test-piece reference number and manufacturing details		
Size of sampled ceramic		
Number of test-pieces		
Test-piece preparation	Method of cutting Method of mounting Method of grinding and polishing Method of etching	
Method of observation	Type of microscope Calibration method Calibrated magnification Method of recording images Method of processing images (manual, automatic)	
Results for each micrograph	Total line length Number of intercepts for each line Mean linear intercept size	
Remarks Problems with preparation Features of microstructure Aspect ratio of grains		
Micrographs		
Signature of responsible person		

Appendix 2 : Instructions for the Round Robin

CEN TC184/WG3 AND VAMAS TWA3

GRAIN SIZE ROUND-ROBIN INSTRUCTIONS

Materials and equipment checklist

This round-robin expects the participants to have:

- Optical or scanning electron microscope with calibration graticules or gratings, and photomicrographic facilities (e.g. Polaroid)
- Metallographic mounting and polishing equipment
- Furnace capable of 1550 °C for short-term thermal etching in air
- Evaporation or sputter coating equipment
- Steel ruler, pair of compasses
- Computer capability for listing grain size data (see Appendix)

The materials supplied by the organisers include:

- These instructions
- Copy of draft standard for grain size measurement by mean linear intercept method
- Two copies of a computer-drawn "microstructure"
- Two specimens of alumina, one brown (Material 1) and one white (Material 2)
- One large micrograph of material 2
- One transparent grid for porosity measurement
- Two transparencies for grain size measurement by mean linear intercept method on computer drawn microstructure
- Two transparencies for grain size measurement by mean linear intercept method on large micrograph of material 2
- One transparency for grain size measurement on large micrograph of material 2
- One reply form

1. INTRODUCTION

The Organisers are grateful for your agreement to participate in this round robin, which is being run via both the CEN Technical Committee 184, Working Group 3 for Advanced Technical Ceramics, and through VAMAS TWA3, Ceramics. When standards are proposed in a new technical area for the first time, there are considerable uncertainties about the range of their applicability or their usability by ordinary research or quality control laboratories. The characterisation of microstructure is no exception. The evaluation of proposed test methods for standardisation in an area such as this can only be performed by round-robin exercises which provide indications of the scatter of results between laboratories and the degree of difficulty experienced. The results will tell us how successful the draft methods are, and will show us where modifications are needed. The starting point is a new CEN draft for a manual grain size measurement based on a "ceramic" version of ASTM E112. The round-robin has been designed to have a series of tests of various aspects of measurement which will allow the organisers to estimate the size of the scatter due to individual participants' approaches to the tasks in relation to the randomness of selection of areas in real samples. An element of difficulty has been incorporated to test limitations. We hope that you will take the measurement seriously. It is intended that the task should take no longer than a few days of a moderately experienced person's time.

2. ROUND-ROBIN OBJECTIVES

1. The primary objective is to test the reproducibility of the procedure in the CEN draft standard for grain size measurement by the manual mean linear intercept method. This is being done in two ways:

   (i) The first way uses an computer-drawn "ideal" microstructure with no porosity and no second phase. This will determine the scatter of results between participants inherent in the random positioning of lines or circles on the "micrograph", which represents a minimum attainable scatter without significant error from misinterpretation of grain boundaries or other features.

   (ii) The second way is practical, and is based on repeating the method on a polished and thermal etched specimen of Material 1. This will determine the additional scatter due to variation between areas chosen randomly on specimens from a single batch of material. A second material with a strongly bimodal grain size is also examined to study the use of the method when the level of scatter is likely to be much higher.

2. The RR also has two secondary objectives which relate to possible future standards, but which can be performed at the same time on the same micrographs:

   (iii) To test a grid method for measuring porosity by counting intersections that lie over pores. The grid size has been chosen to be a compromise between being large enough for visual counting, yet small enough to be of the same scale as the features to be counted.

   (iv) To test a draft procedure for manually producing a grain size distribution

To assist in the analysis of the results, we ask you to draw lines and grids on supplied transparencies used to overlay micrographs, rather than drawing lines on the micrographs themselves, so that comparisons are more easily effected.

3. NOTE ON AUTOMATIC IMAGE ANALYSIS METHODS

The organisers of this round-robin are fully aware that many industrial companies and institutes have access to automatic image analysis sytems, and that the development of standards should reflect this. In the draft CEN standard it has not proved possible to allow for such methods because of uncertainties in the methods employed and there lacks an intercomparison of manual and automatic methods. The principal objectives of this round-robin are to examine manual methods as defined in the standard. However, if participants who have automatic methods are willing to employ them **in addition** to the manual methods on the same micrographs, the results would provide a very welcome comparison. Please use the relevant parts of the report form to describe the procedures adopted, and in particular the method by which the system calculates the parameters where this is defined in documentation.

4. ROUND ROBIN TESTS

PART 1 - COMPUTER-DRAWN MICROSTRUCTURE

Using the text of the standard as the method, fix one of the supplied computer-drawn "microstructures" to a board or a desk, and overlay the transparency to exactly match the borders. Draw at least five lines at random angles and positions, and of length greater than 75 mm, on the transparency using a thin pen. Count the number of intersections of the lines with the "grain boundaries". Use the rules in the standard for the case where the line intersects a triple point. Measure the total line length to the nearest millimetre, and calculate the "grain size" in millimetres. Report on the attached report sheet.

Replace the transparency with the second one, overlaying to exactly match the borders. Draw on the transparency at least three randomly positioned circles of greater than a minimum diameter to be determined according to the instruction in the draft standard. Follow the procedure in the draft to count the number of grains. Measure the diameters of the circles to the nearest millimetre. Compute the total circumference of all the circles and calculate the mean "grain size" in millimetres. Report on the results sheet.

PART 2 - MATERIAL 1

Take the supplied specimen of material 1, mount it in mounting medium and grind and polish to obtain a scatch-free surface with a minimum of grain pluck-out. This material has an equiaxed grain structure, and polishes reasonably well. Remove the specimen from the mount and thermally etch it to a level where the grain boundaries are distinctly visible in a micrograph. A temperature of about 1350 °C for 30 minutes should suffice.

After applying a metallic coating as necessary, take two identical high-magnification micrographs of at least 500X, but 1000 - 1500X would be better, using an optical microscope or SEM. Use certified graticules to calibrate the magnification of the optical microscope, and a calibrated grating or spheres to calibrate the SEM in both horizontal and vertical screen

directions. Use the procedure for determining acceptability of uniformity of magnification as described in the draft standard. Report the calibration on the report sheet.

Use the procedure described in the draft standard for determining the mean grain size by both the line and the circle methods, this time drawing directly on the micrographs. Report the total line length in both cases, the numbers of grains crossed, and the mean linear intercept grain sizes calculated in micrometres.

PART 3 - MICROGRAPH

The supplied large micrograph is of a randomly selected area of material 2, a high-purity material, and shows a distinct bimodal grain size distribution. The contrast and grey level have been adjusted to minimise pore edge brightness without obscuring thermal etch boundaries.

First, use the supplied transparent grid to measure apparent porosity of the **entire area** of the micrograph. The grid size has been chosen to provide sufficient frequency of counting to ensure that most pores should be intersected. Pin or tape the micrograph to a board or other suitable surface, overlay the grid in a random position, but fully covering the micrograph, and tape it down. Count the number of intersections of the grid that lie over the micrograph. Count the number of intersections that lie over obvious pores. If, as far as you can judge, the grid lies exactly on a pore boundary, count this as half an intersection. Divide the number of intersections that lie over pores by the total number over the field area of the micrograph, and report the result as a percentage. The example grid that has been supplied is probably the minimum size that can be used without excessive eye-strain. To minimise eye-strain use good illumination and count each horizontal grid line in turn, masking off conflicting information from other grid lines with sheets of paper.

Second, use the line and/or the circle method in the draft standard to calculate the mean linear intercept grain size. As in Part 1, use the supplied transparencies overlaid on the micrograph, one for lines and the other for circles. Report the result in millimetres on the micrograph.

Third, determine a grain size distribution according to the procedure in Annex 1. This exercise is intended to examine the possibilities for developing a standard along these lines, and by using the same micrograph, the likely spread of apparent distributions obtained on the same area by the various participants can be determined. Report the results on the report sheet, and preferably as an ASCII file on a floppy disc.

PART 4 - MATERIAL 2

Prepare a polished and thermally etched surface from the supplied piece of Material 2. This material is not easy to polish, and this part of the RR is designed to examine the influence that difficulties in surface preparation has on the results obtained for porosity measurement, as well as the scatter in mean grain sizes resulting from choice of area for analysis. It is recommended that particular care is taken **not** to use compliant laps, and to avoid grinding with coarse grits if possible, and to employ sufficiently long polishing times with grit sizes less than 6 μm to ensure removal of pluck-out damage due to prior coarse grinding. Some

repeat attempts to polish may be required until a minimum apparent pluck-out is obtained by optical inspection. The example micrograph supplied was set in bakelite mounting medium and was polished directly with 6 μm diamond grit on stainless steel mesh with no prior grinding to minimise pluck-out.

Prepare at least two identical micrographs, typically of the magnification of that of the supplied micrograph, of what you feel is a representative area, using optical microscopy or SEM. Avoid obvious areas of pluck-out. Use the procedure in the draft standard to obtain mean linear intercept grain size by drawing lines and/or circles on separate micrographs. Use the procedure in Annex 1 to determine the grain size distribution.

5. REPORT

The attached report form should be completed as fully as possible, and should be returned as indicated together with **all** items listed. This is important because the evaluators may need to check on the individual's employed technique.

R Morrell
National Physical Laboratory
Teddington, UK

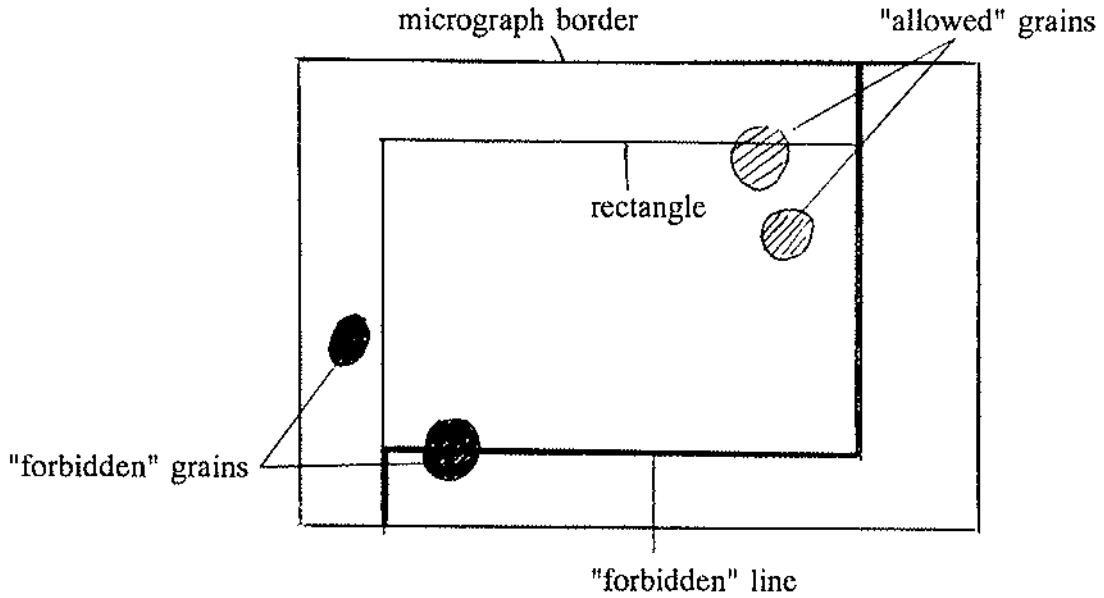
14 February 1992
CENGSRR.TXT

APPENDIX 1 - METHOD FOR DETERMINING GRAIN SIZE DISTRIBUTION

This method is the practical part of a proposal for determining manually a grain size distribution. The steps are as follows, and should be interpreted to suit the micrograph concerned. Participants are not asked to determine the actual distribution themselves as part of the exercise; the project coordinators will do this, and request that the data are supplied on floppy disc for convenience.

1. Draw on a transparency a rectangle parallel to the sides of the micrograph. The rectangle should be as large as possible, but such that the boundary of the rectangle intersects only grains that are fully visible in the micrograph. As a guide, there should be a region of approximately 1.5 x the mean largest grain size gap between the rectangle and the edges of the micrograph.

2. Mark two adjacent sides of the rectangle as "forbidden lines". Include in the analysis all the grains that lie within the rectangle ("allowed" grains), excluding those intersecting the forbidden line ("forbidden" grains), but including all those that intersect the other two sides of the rectangle (see Figure).



3. Draw a uniform grid over the entire area of the micrograph, including the border region. Select the spacing of the grid to be approximately the same size as the largest grains so that each of these are not normally counted twice. (NOTE: If this would require less than 10 grid lines to cover the horizontal direction in the micrograph, the magnification of the micrograph is too large for effective analysis, and would give poor statistics.)

4. Use **either** Method 1 **or** Methods 1 **and** 2 as below:

Method 1: Measure the lengths in millimetres to the nearest 0.5 mm of the distances between intercepts of grain boundaries with each horizontal and vertical grid line of "allowed" grains (see Figure above) based on the rectangular area.

Method 2: Measure the lengths in millimetres to the nearest 0.5 mm of the distances between intercepts of grain boundaries with each horizontal and vertical grid line of all grains that lie **wholly within the area of the micrograph**, i.e. ignoring the drawn rectangle drawn for method 1 and also any grains which intersect the edges of the micrograph.

Note: Method 1 is the classical approach which removes bias. Method 2 has bias that can be removed mathematically, and we wish to test both. The measurements are best listed first for method 1, and then the **additional** intercept methods for method 2 are added in separate columns.

5. Calculate the mean and standard deviation of the measured intercept lengths in millimetres. If the magnification of the micrograph is known, calculate the mean and standard deviation also in micrometres.

Note: We have not asked you to draw of grain size distribution of your own, but obviously you can if you wish. We suggest two possible methods of assessing the individual measurements:

1. Inspect the measurements for the highest and lowest values. Choose a round number size less than the smallest and greater than the largest. Divide the range of values into, say, 10 intervals of equal logarithmic size, e.g. 1 to 1.5, 1.5 to 2.25, 2.25 to 3.38, 3.38 to 5.07, etc. (factors of 1.5 each time). Plot a histogram of the numbers of intercept distances falling into each category of interval.
2. Rank the intercept values in ascending order of size and plot cumulative probability versus log(intercept size).

6. The report shall include:

- the transparency with the rectangle and grid drawn upon it
- the calculated mean and standard deviation of the intercept lengths measured
- an ASCII file on a 3.5" floppy disc (formatted to 720 kb under DOS3.3 or higher) containing:

(line 1)	the number of measured intercepts
(subsequent lines)	the individual length values in millimetres (method 1)
(line space)	
(line)	the additional number of intercepts in method 2 if performed
(subsequent lines)	the individual length values in millimetres (method 2)

RESULTS FORM - GRAIN SIZE ROUND ROBIN

When the exercise is complete, please return this form, together with the following items:

- Two transparencies for grain size measurement on the computer drawn microstructures, one with lines and one with circles drawn (Part 1);
- Two identical micrographs of Material 1 (brown) prepared by the participant with lines and with circles drawn (Part 2);
- Two transparencies for grain size measurement on the supplied large micrograph of material 2, one with lines and one with circles drawn (Part 3);
- One transparency for grain size distribution measurement on supplied large micrograph of material 2 with rectangle and grid drawn (Part 3);
- Micrographs of Material 2 (white) prepared by the participant with lines and/or circles drawn, and with the rectangle and grid drawn for grain size distribution (Part 4);
- Floppy disc containing grain size distribution measurements.

Please return as soon as possible to:

Dr L Dortmans
Centre for Technical Ceramics
University of Eindhoven
PO Box 513
5600 MB EINDHOVEN
Netherlands

Phone (+31) 40 472770

Fax (+31) 40 445619

If there are any questions during the exercise, please contact either Dr Dortmans, or Dr R Morrell, National Physical Laboratory, Teddington, Middlesex TW11 0LW, UK, Phone (+44) 81 943 6381 or Fax (+44) 81 943 2989.

Participant's details:

Name..... Date of tests.....

Address.....

.....

.....

.....

Phone..... Fax.....

Type of optical microscope.....

Type of scanning electron microscope.....

Type of coating used on test specimens.....

Optional automatic image analysis:

Equipment type.....

.....

Software package used.....

PART 1 - COMPUTER-DRAWN MICROSTRUCTURE

Line method:

Total length of straight lines .....mm

Number of counted intersections .....

Mean linear intercept size .....mm

Circle method:

Diameters of drawn circles .....mm .....mm .....mm

Number of intersections .....

Mean linear intercept size .....

Optional automatic image analysis:

Scanning method horizontal? vertical? both? random directions?

Mean linear intercept size .....mm

PART 2 - MATERIAL 1

Polishing:

Polishing method used .....

.....

Thermal etch time/temperature .....

Microscope:

Nominal magnification of microscope .....

Calibrated magnification from graticule or grid:

Optical microscope (M) .....

Scanning electron microscope:

Horizontal (M_h) .....

Vertical (M_v) .....

Mean value ($M = 0.5(M_h + M_v)$) .....

Line method:

Total line length .....mm

Total number of intersections .....

Mean linear intercept size .....μm

Circle method:

Diameters of drawn circles .....mm .....mm .....mm

Total circle circumference .....mm

Number of intersections .....

Mean linear intercept size .....μm

Optional automatic image analysis:

Method (if different from above).....

Mean linear intercept size .....μm

PART 3 - MICROGRAPH

Transparent grid porosity determination:

Number of intersections of grid over micrograph .....

Number of intersections of grid over pores .....

Area fraction of porosity .....%

Optional automatic image analysis:

Analysis method used.....

.....

Area fraction of pores .....%

Grain size, line method:

Lengths of lines drawn .....mm .....mm .....mm .....mm

.....mm

Total line length .....mm

Total number of intersections .....

Mean linear intercept size .....mm

Grain size, circle method:

Diameters of drawn circles .....mm .....mm .....mm

Total circle circumference .....mm

Total number of intersections .....

Mean linear intercept size .....mm

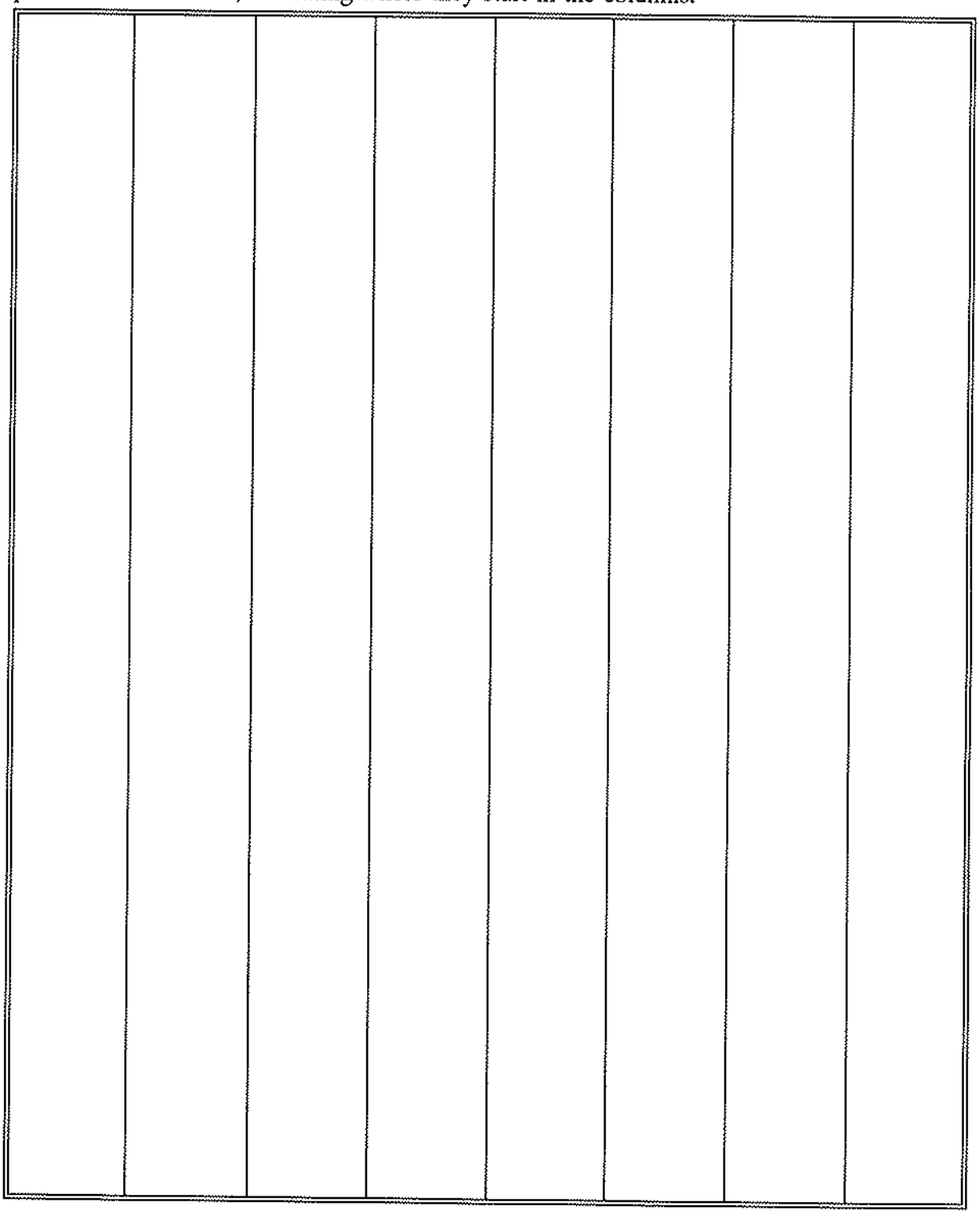
Optional automatic image analysis:

Mean linear intercept size .....mm

PART 3 - MICROGRAPH (cont.)

Grain size distribution:

Give the values of intercept distances to the nearest 0.5 mm along each grid line in the for all "allowed" grains (method 1) in order of measurement (filling the columns in turn). If you are willing to extend the task to incorporate the additional intercept distances for method 2, please add these also, indicating where they start in the columns.



PART 3 - MICROGRAPH, grain size distribution (cont.)

	Method 1	Method 2
Total number of intercepts	.....	.....
Mean intercept length, mm	.....	.....
Standard deviation, mm	.....	.....

Optional automatic image analysis:

Method of analysis employed.....

.....

.....

.....

Mean grain size .....mm

Please enclose table of data and plot of results

PART 4 - MATERIAL 2

Polishing:

Polishing method used .....

.....

Thermal etch time/temperature .....

Microscope:

Nominal magnification of microscope .....

Calibrated magnification from graticule or grid:

Optical microscope (M) .....

Scanning electron microscope:

Horizontal (M_h) .....

Vertical (M_v) .....

Mean value ($M = 0.5(M_h + M_v)$ .....

Line method:

Total line length .....mm

Total number of intersections .....

Mean linear intercept size .....μm

Circle method:

Diameters of drawn circles .....mm .....mm .....mm

Total circle circumference .....mm

Number of intersections .....

Mean linear intercept size .....μm

Optional automatic image analysis:

Method (if different from above).....

Mean grain size .....μm

PART 4 - MATERIAL 2 (cont.)

Give the values of intercept distances to the nearest 0.5 mm along each grid line in the for all "allowed" grains (method 1) in order of measurement (filling the columns in turn). If you are willing to extend the task to incorporate the additional intercept distances for method 2, please add these also, indicating where they start in the columns.

PART 4 - MATERIAL 2, grain size distribution (cont.)

	Method 1	Method 2
Total number of intercepts	.....	.....
Mean intercept length, mm	.....	.....
Standard deviation, mm	.....	.....
Magnification, X	.....	.....
Mean intercept length, μm	.....	.....
Standard deviation, μm	.....	.....

Optional automatic image analysis:

Method of analysis employed.....

.....

.....

.....

Mean grain size, μm .....

Standard deviation, μm .....

Please enclose table of data and plot of results.

COMMENTS

Please comment here about any difficulties experienced with any of the methods within this round-robin.

Appendix 3 : Preparation procedures for material 1

This Appendix gives an overview of the procedures applied to obtain a micrograph of the sample of material 1 by the various participants.

P1:
Polishing: diamond paste 40, 20 and 10 micrometre. Pb disk 1 micrometre d.p.
Etching: 1350 C, 10 min

P2:
Polishing: polishing with 6 micrometre grit (5 min) and 1 micrometre grit (65 min)
Etching: 1350 C, 30 min

P3:
Polishing: BORCARBID 320 from 20 to 50 micrometre, 10 min. BORCARBID 800 from 3 to 9 micrometre, 10 min. Diamond 2 micrometre, 10 min. and diamond 0.5 micrometre for 10 min.
Etching: 1400 C, 30 min

P4:
Polishing: SiC papers grit 600/10 min, grit 800/15 min, grit 1200 15 min.. Diamond polishing on napless cloth 15 micrometre/120 min, 9 micrometre/90 min, 3 micrometre/90 min, 1 micrometre/30 min.
Etching: 1350 C, 30 min

P5:
Polishing:
Etching: 1300 C, 30 min

P6:
Polishing: diamond discs 125, 46, 25 and 15 micrometre. Polishing with 8, 6 3, and 1 micrometre hard cloth
Etching: 1350 C, 30 min

P7:
Polishing: 63 micrometre/10 min, 30 micrometre/30 min, 10 micrometre/20 min, 3 micrometre/25 min, 0.25 micrometre/30 min.
Etching: 1350 C, 5 min

P8:
Polishing: SiC paper to 1200 grit, 6 micrometre diamond paste on tin lap 15 min, 6, 1 and 0.5 diamond spray on PSU cloth, OP-S solution napped cloth for 2 minutes
Etching: 1350 C, 10 min

P9:
Polishing: grinding with 20 micrometre hard diamond lap, grinding with 6 micrometre spray on metal based lap (20 min), polishing with diluted 6 micrometre diamond paste on nylon cloth (20 min), polishing with diluted 3 micrometre

diamond paste on nylon cloth (40 min), polishing with diluted 1 micrometre diamond paste on silk cloth (20 min) and polishing with diluted 0.25 micrometre diamond paste on silk cloth (20 minutes)

Etching: 1200 C, 30 minutes

P10:

Polishing: 20 micrometre diamond disc on hard composite lap, 6 micrometre diamond spray on a hard metallic lap (20 min), 6 micrometre diamond paste on a hard napless cloth (20 min), 3 micrometre diamond paste on a hard napless cloth (40 min), 1 micrometre diamond paste on a hard napless cloth (20 min) and 0.25 micrometre diamond paste on hard napless cloth (10 min)

Etching: 1200 C, 30 min

P11:

Polishing: 63-74 micrometre SiC to level. 15 micrometre SiC until scratches removed. 6 and 3 micrometre spray on silk until scratches removed

Etching: 1250 C, 60 min

P12:

Polishing: diamond grinding grit 600, 2 MPa, 3 min.. Petrodisc 6 micrometre, 8 min.. Pan (DP Mat) 6 micrometre, 1.5 MPa, 20 min.. Pan-W 3 micrometre, 1.5 MPa, 20 min.. DP-Plus 1 micrometre, 1.5 MPa, 20 min.

Etching: 1350 C, 10 min

P13:

Polishing: diamond disc grains 15 micrometre for 30 min., diamond disc grains 6 micrometre for 20 min., cloth with diamond paste 1 micrometre for 25 min.

Etching: 1350 C, 15 min

P14:

Polishing: 6 micrometre diamond on soft cloth; 3 and 1 micrometre diamond on hard cloth.

Etching: 1350 C, 15 min

P30:

Polishing: diamond grinding discs 115, 65 and 20 micrometre; Cu-Fe lapping wheel 6 micrometre; DP-DAC cloth 6 3 and 1 micrometre diamond; DP+ cloth on vibratory mill 0.5 micrometre diamond

Etching: 1350 C, 30 min

P31:

Polishing: Buehler Automat IV, 150 rpm. 45, 15, 3 and 1 micrometre diamond powder on rayon pad

Etching: 1100 C, 30 min

P32:
Polishing: diamond paste on nylon cloth 30, 15, 9, 3 and 1 micrometre, 10 min/step, 10 psi, 300 rpm
Etching: 1250 C, 15 min

P33:
Polishing: 1 micrometre diamond lapping 4 hours
Etching: 1350 C, 30 min

P34:
Polishing: lap wheel 30 micrometre, lap wheel 10 micrometre, silk cloth 3 micrometre, paper cloth colloidal SiO2
Etching: 1350 C, 30 min

P35:
Polishing: Buehler Dialog sample preparation. 30 micrometre diamond 5 min, 6 micrometre diamond 20 min, 1 micrometre diamond 20 min
Etching: 1350 C, 30 min

P36:
Polishing: 240,320,400,600 SiC papers; 6-12 micrometre diamond paste w/kerosene on lead lap for 8-16 hrs; 0.05 micrometre CeO in water on pellon cloth for 8 hrs; 0.05 silica in water on nylon cloth for 16-24 hrs
Etching: 1200 C, 20 min

P37:
Polishing: 30 micrometre diamond film on mylar 4 minutes; 15 micrometre diamond paste on polishing cloth 10 minutes, 400 rpm; 6 micrometre diamond paste on polishing cloth 10 minutes, 400 rpm; 1 micrometre diamond paste on polsihing cloth 8 minutes, 400 rpm; 0.25 micrometre diamond paste on polishing cloth 4 minutes, 400 rpm; colloidal silica (0.05 micrometre) on polishing cloth 4 minutes 300 rpm.
Etching: 1350 C, 6 min

P38:
Polishing: 1 micrometre diamond grains and polishing cloth on rotating disk
Etching: 1350 C, 30 min

P39:
Polishing: diamond polishing
Etching: 1350 C, 30 min

P40:
Polishing: 15 micrometre diamond disk, 2 and 0.25 micrometre diamond on cloth
Etching: 1350 C, 30 min

Appendix 4 : Preparation procedures for material 2

This Appendix gives an overview of the procedures applied to obtain a micrograph of the sample of material 2 by the various participants.

P1:
Polishing: as for material 1
Etching: 1350 C 10 min, 1400 C 10 min, 1550 C 15 min

P2:
Polishing: not specified
Etching: 1650 C, 15 min

P3:
Polishing: as for material 1
Etching: 1500 C, 30 min

P4:
Polishing: as for material 1
Etching: 1500 C, 120 min

P5:
Polishing:
Etching: 1500 C, 30 min

P6:
Polishing: as for material 1
Etching: 1450 C, 120 min

P7:
Polishing: 40 micrometre/10 min, 30 micrometre/20 min, 10 micrometre/30 min, 3 micrometre/60 min, 0.25 micrometre/60 min. Cloth type: PA-W, 50 rpm/2.5 bar
Etching: 1500 C, 60 min

P8:
Polishing: 1200 grit SiC, 6 micrometre diamond paste on tin lap, 6 micrometre diamond spray on PSU cloth for 40 min, 1 micrometre diamond spray on PSU cloth for 90 min, 0.25 micrometre diamond spray on PSU for 60 min, OP-S solution napped cloth for 1 minute
Etching: 1350 C, 30 min

P9:
Polishing: grinding with 20 micrometre hard diamond lap, grinding with 6 micrometre spray on metal based lap (20 min), polishing with diluted 6 micrometre diamond paste on nylon cloth (20 min), polishing with diluted 3 micrometre diamond paste on nylon cloth (20 min), polishing with diluted 1 micrometre diamond paste on silk cloth (20 min) and polishing with diluted 0.25

micrometre diamond paste on silk cloth (20 minutes)
Etching: 1400 C, 60 min

P10:
Polishing: 20 micrometre diamond disc on hard composite lap, 6 micrometre diamond spray on a hard metallic lap (10 min), 6 micrometre diamond paste on a hard napless cloth (20 min), 3 micrometre diamond paste on a hard napless cloth (20 min), 1 micrometre diamond paste on a hard napless cloth (20 min) and 0.25 micrometre diamond paste on hard napless cloth (5 min)
Etching: 1400 C, 60 min

P11:
Polishing: 15 micrometre SiC to level. 6 micrometre SiC until scratches removed and minimal pullout. Then 3 micrometre SiC until scratches removed and minimal pullout
Etching: 1500 C, 60 min

P12:
Polishing: as for material 1
Etching: 1350 C, 30 min (Comment from P12: etching temperature possibly was too low, but the furnace broke down so no further etching could be done)

P13:
Polishing: as for material 1
Etching: 1350 C, 45 min

P14:
Polishing: 6 micrometre diamond on soft cloth; 3, 1 and 0.25 micrometre diamond on hard cloth.
Etching: 1400 C, 90 min

P30:
Polishing: as for material 1
Etching: 1500 C, 30 min

P31:
Polishing: as for material 1
Etching: 1500 C, 120 min

P32:
Polishing: as for material 1
Etching: 1450 C, 15 min

P33:
Polishing: as for material 1
Etching: 1380 C, 60 min

P34:
Polishing: lap wheel 30 micrometre, lap wheel 6 micrometre, silk cloth 3 micrometre, 3 micrometre paste+colloidal SiO2 on paper cloth, colloidal SiO2
Etching: 1350 C, 30 min

P35:
Polishing: as for material 1
Etching: 1400 C, 30 min

P36:
Polishing: as for material 1
Etching; 1200 C, 30 min

P37:
Polishing: as for material 1
Etching: 1450 C, 12 min

P38:
Polishing: as for material 1
Etching: 1500 C, 60 min

P39:
Polishing: as for material 1
Etching: 1350 C, 30 min

P40:
Polishing: 2 micrometre diamond paste; 0.25 micrometre diamond paste
Etching: 1500 C, 30 min