

REPORT

Measurement of WC Grain Size Distribution

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ABSTRACT

A variety of measurement methods to characterise the WC grain size and distribution of WC/Co hardmetals has been examined. Computer simulations from model structures were used to provide support to the measurement methods. In addition, the shape of WC grains in 3-D was investigated by sequential polishing of a WC/Co hardmetal followed by reconstruction of selected grains.

It is recommended that size distributions of area plotted against size or normalised size provide the most effective method for discriminating between different grain size distributions. An estimate of area can be used indirectly by squaring intercept values rather than making direct measurements of grain areas. A representative mean value for the WC grain size should be expressed as an arithmetic number average.

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Approved on behalf of Managing Director, NPL, by Dr C Lea,
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1 INTRODUCTION

1.1 INDUSTRIAL NEED

The mechanical properties of hardmetals, like many other engineering materials, are strongly influenced by their grain size [1,2]. However, for hardmetals there is no standard method for measuring the size of the WC grains or their distribution. Lack of reference methods for this purpose is hindering a fundamental understanding of the link between grain size, distribution and mechanical properties. In particular, a knowledge of how measurements in two dimensions are related to a three dimensional shape distribution is vital if the question of how uniformity (or non-uniformity) of structure relates to strength and toughness.

It is sometimes claimed that more uniform structures have performance advantages while other claims report that a non-uniform structure has benefits. The lack of an agreed method to measure the extent of uniformity is clearly a hindrance to optimised use. At present subjective assessments are made from an evaluation by eye of structural images.

1.2 QUALITY CONTROL

Composition is conventionally specified in hardmetal in terms of how much Co is present in wt% and a nominal figure for the grain-size of the WC. In the sintered material the binder-phase is in fact an alloy of Co containing W and C in solid solution. The amount of alloying elements present can be determined by measurements of magnetic properties such as coercivity and saturation magnetisation and methods exist to measure these properties. However, measurement of the WC grain-size is not so straightforward. There is no accepted NDT method for measurement of this parameter, although coercivity is sometimes used empirically. The coercive force required to demagnetise a WC/Co hardmetal is related to the Co/WC interphase area, because domain walls are pinned by these phase-boundaries. The Co/WC interphase area is inversely related to the WC grain-size. For smaller values of the latter the interphase area increases and consequently so does the coercivity. However, coercivity also changes with change in Co binder-phase composition and this should be allowed for in empirical equations relating WC grain-size to coercivity.

Internationally, for the purpose of grade specification, there is as yet no widely agreed method for measurement of WC grain-size, although in the research community it is fairly common practice to use the linear intercept technique. The intercept method has the advantage of providing information on the size distribution of the WC grains as well as a mean value.

In recent work at NPL supported by the British Hardmetal Association [3] the grain size distributions of about fifty samples of mostly commercial, 6 wt% Co WC-based hardmetals were obtained using the linear intercept method. The grain size distributions were markedly similar considering the wide range of material source and processing routes. However, the intercept method did not appear to discriminate very well between materials, which, to the eye, had subjectively different size distributions of grains. Thus, for distribution measurement there is a need for developing methods which are more discriminating.

1.3 MICROSTRUCTURAL METROLOGY

The most direct way of measuring the WC grain-size is to polish and etch a cross-section of the microstructure and then to use quantitative metallographic techniques to measure a mean value for the grain-size either by point or area counting or by linear intercept techniques. This can be performed using optical microscopes for some coarser-grained WC hardmetals but for many materials the grain-size is too fine to be resolved optically and so scanning electron microscopes (SEM) must be used. In some ultra-fine grained hardmetals the average grain size is very small, less than 0.5 μm , and good quality images are difficult to obtain in

conventional SEMs. Thus a high resolution instrument using a field emission electron beam source is required. A measurement good practice guide which addresses the various techniques for good image preparation is currently being written at NPL [4].

The linear intercept technique is popular because it produces size distribution information. It is often found that the measured intercept distribution is lognormal in form, which gives a straight line when plotted on lognormal probability paper [5]. Another option is measurement of the sectional grain area, however, this is not yet commonly used or well developed. However, as an alternative the intercept values can be squared to give an estimate of area and hence area (size) distribution.

1.4 WC SIZE DISTRIBUTION

The effect of size distribution as well as mean size of WC grains on hardmetal properties has not been extensively researched although widespread views are held on the effects of distribution width on properties. It is straightforward to ask the question "For the same mean value of grain size are better properties obtained by having a narrow or a wide distribution?". At present there are no published results which allow an unambiguous answer to be quoted. Furthermore, there is the additional and related effect of modality. For example, in principle a structure can be imagined in which there are mixed populations of grains with different means and spreads. The effects of such a structure on properties are not known, although at one extreme a material structure can be envisaged in which a few isolated extremely large grains are dispersed within a uniformly distributed structure. If, for example, these grains are larger than about 20-30 μm in a nominally 1 μm mean-sized material then it has been shown [6] that, when subjected to tensile stresses, they will act as fracture initiation sites, and thus have an obvious effect on properties. But for less extreme cases such as, for example, a 50/50 or 75/25 bimodal distribution, the effect on properties is less clear.

1.5 MEASUREMENTS AND COMPUTER SIMULATION

In this work a comparison has been made of measurements made directly on two hardmetals with studies of size distribution obtained by computer simulation. Both conventional WC/Co hardmetals (codes D1 and M15, i.e. 6 and 15 wt%Co hardmetals, respectively) and a model material with a wider WC grain size distribution than normal (code F12) were examined. Two computer simulation approaches were used, one based on the use of a transputer-based method for producing simulated images of hardmetal structures and the other based on the use of a PC program (called Polychop) that enables random slices and intercepts to be drawn through a number of different shapes, including a cube, cylinder, tetrahedron, tetrakaidecahedron and trigonal prism. The computer studies were complemented by measurements of WC grains obtained experimentally by sequential polishing of a coarse grained WC/11wt% Co hardmetal followed by reconstruction of selected grains.

2 SYSTEMATIC DISTRIBUTION ANALYSIS

2.1 DISTRIBUTION TYPES

Intercept data can be plotted systematically by a number of methods in order to compare and contrast the different ways in which distributions of sizes can be displayed and examined [3], in particular by using distributions based on either number or size probabilities.

For example:

- the data can be shown as cumulative probability plots with the ordinate (y) given a linear scale or by using probability paper. The latter method expands the extremes of the distribution and contracts the central regimes.
- the data can be plotted as cumulative probability plots with the abscissa (x) on a linear or logarithmic scale.
- a further refinement is to normalise the abscissa values, ie normalised intercept or normalised area. This is obtained by dividing by the number or size average.
- the data can be plotted as intercepts, or areas, either by number (A) of the i^{th} ranked data plot or as cumulative length of intercept (B) or cumulative area (C). Area can be estimated by squaring the intercepts. For A, B and C the mathematical expressions used to calculate cumulative probability, P_i are given by

Number Distribution:

$$P_i = \frac{\sum_{i=1}^i n_i - 0.5}{N_d}, \quad (\text{A) for intercepts} \quad (1)$$

$$P_i = \frac{\sum_{i=1}^i n_i - 0.5}{N_A}, \quad (\text{A) for areas or estimated areas} \quad (2)$$

Size Distribution:

$$P_i = \frac{\sum_{i=1}^i d_i}{\sum_{i=1}^i d_i}, \quad (\text{B) for intercepts} \quad (3)$$

$$P_i = \frac{\sum_{i=1}^i A_i}{\sum_{i=1}^i A_i}, \quad (\text{C) for areas or estimated areas} \quad (4)$$

where N_d is the total number of intercepts, d_i , counted

N_A is the total number of grain areas, A_i , counted

If area is estimated from intercepts squared then $N_d = N_A$

$\sum_{i=1}^i n_i$ is the cumulative number of intercepts or areas up to the i th data point

$\sum_{i=1}^i d_i$ is the cumulative length of intercept up to the i th data point

$\sum_{i=1}^N d_i$ is the total cumulative length of intercept

$\sum_{i=1}^i A_i$ is the cumulative area up to the i th data point

$\sum_{i=1}^N A_i$ is the total cumulative area.

Some of the different distribution types are illustrated in Fig 1.

There are several questions to be asked of these different types of data plots.

- *Are differences revealed more clearly when data is plotted using a linear abscissa rather than logarithmically? Are there advantages in using normalised data for the intercept values?*

When the data are plotted against intercept or area on a linear abscissa the distribution is compressed for the small sizes and expanded for the large sizes. This representation possibly reflects what is seen when the eye/brain interprets the microstructures with a bias towards the larger sizes. It enables differences to be seen more easily for the large WC grains but not for the WC grains at the small end of the distribution. In conclusion, both plots could have a role to play depending on the kind of information required. Normalised abscissa values have some advantage when comparing materials with different mean values. It is possible to more clearly see differences in distribution widths.

- *Is there any benefit in plotting the ordinate on a linear scale rather than on probability paper?*

The probability plots expand the distribution at each end and effectively weight the information so that perhaps an exaggerated effect of differences in large or small grains can be observed.

- *Is there an advantage to plot the data as distance or area fractions (ie size fraction) rather than by number fraction? Is it number fractions or is it size fractions that are related to properties?*

It has been shown [3] that there are advantages to using areas rather than intercepts because the differences in distribution type for materials with narrow and wide distributions are more pronounced. Areas can be estimated by squaring the values of the intercepts (see section 3.1). Graphs of areas or estimated areas plotted against normalised size appear to give the best discrimination between different distributions.

2.2 PROPERTY/DISTRIBUTION DEPENDENCIES

The type of distribution measurement to be preferred depends on what effect a specific distribution of sizes has on properties. At present it is not known whether the properties are determined by the number fraction of grains of a given size or by length fraction, by area fraction, or by volume fraction.

Material structures are generally characterised, apart from their chemical composition, by a size parameter, S . In single phase materials the most commonly used size parameter is grain size, d , which has the dimensions of length. It is common practice to measure grain size by the length of intercept, d , within each grain of a line superimposed across the image of the microstructure because structures are usually measured on two-dimensional polished surface. Many intercepts are measured on random lines and the number average is taken to give an arithmetic mean linear intercept, d_m .

$$d_m = \frac{\sum d_i}{N} \quad (5)$$

where N is the total number of intercepts and d_i are individual intercept lengths. This value in many cases is taken to be the grain size without converting it to a true 3-D grain diameter, which requires the use of a multiplication factor dependent on the shape of the individual grains.

Property models are generally related to the size parameter, S , by equations of the type

$$P = a + bS^n \quad (6)$$

where a , b and n are constants and P is the property of interest. For example in hardmetals the Hall-Petch equation can be used to predict the effect of grain size, d , on hardness, H , with

$$H = H_0 + E_1 d^{-1/2} \quad (7)$$

Thus, expression (7) shows by comparison with expression (6) that $a = H_0$, $b = E_1$ and $n = -1/2$.

A model for the toughness, G , can be simply related to the grain size of the cobalt phase, d_{Co} , by

$$G = A_1 + B_1 F_{Co} d_{Co} \quad (8)$$

where F_{Co} is the Co binder-phase strength. Thus showing by comparison with expression (6) that $a = A_1$, $b = B_1 F_{Co}$ and $n = 1$.

Also magnetic coercivity, K , is related to grain size of the carbide phase d_{WC} , by

$$K = A_2 + B_2 d_{WC}^{-1} \quad (9)$$

Thus showing by comparison with expression (6) that $a = A_2$, $b = B_2$ and $n = 1$.

Coercivity is in practice related to d_{Co} not d_{WC} , but for conventional hardmetals d_{WC} is proportional to d_{Co} for constant binder-phase volume fraction [7].

When models of these types are compared with microstructural measurements the grain size of the WC or Co, d_{WC} or d_{Co} , is frequently taken to be the arithmetic (number average) mean linear intercept, d_m , obtained from a two-dimensional image and is not the true 3-D grain diameter. Because all single-phased materials have microstructures in which the grains are distributed in size, a more general form for expression (6) can be derived. The structures of single-phased materials can be characterised in several ways other than by using a number distribution of d_i . For example, the grain area or volume distributions can be obtained on a number basis, or by line, by area fraction or by volume fraction. All these different methods of structure characterisation can have different consequences for the predictive models described above since they lead to different integrals which have to be evaluated.

The most common technique is to use a number distribution of size $f_n(S)$; this is frequently found to be lognormal for intercepts (size). However, a different distribution can be developed based on the size fraction $f_s(S)$ rather than number. Size fraction is obtained by dividing the size of interest by the total size and the size fraction distribution is derived by plotting the size fraction against size.

Thus, there are six possible distributions, three of number and three of size, based on intercept, area or volume. These distribution functions are

Number - $f_n(I)$, $f_n(A)$ and $f_n(V)$

Size - $f_s(I)$, $f_s(A)$ and $f_s(V)$

Some of these distributions (ie $f_n(I)$ and $f_n(A)$) can be measured directly but some types of distribution, ie $f_n(V)$ and $f_s(V)$, must be calculated from directly measured distributions, usually by making assumptions about grain shape. Measurements on the grain size of WC in WC/Co hardmetals have shown that all the first four types of distribution (based on intercept or area) generally follow a lognormal pattern, [3,5-9].

The lognormal distribution function has been used [10] to examine the effects of changes in the microstructural phase size distribution on properties such as hardness, coercivity and toughness. Predictions were made for how these properties were changed with differences in distribution width, based on the use of simple model relations between the property of interest and phase size. Both narrower and wider distributions than those found in conventionally processed hardmetals were examined. For coercivity and hardness the change in property depended on whether number (corresponding to an increase in property) or size (corresponding to a decrease in property) were used as distribution weighting operators. For toughness the property increased with size as the distribution weighting operator and was relatively unchanged for number.

These predictions indicate that it is important to reach common agreement on good practice for the measurement of grain size distributions. It may be that different representations are required dependent on the use to which the data are to be put.

3 RESULTS AND DISCUSSION

3.1 WC/Co MICROSTRUCTURE

The microstructures of conventional 6 wt% Co and 15 wt% Co/WC hardmetals (coded D1 and M15 - supplied by Marshalls Hardmetals Ltd) were analysed using both a linear intercept and area measurement technique to provide data on distribution width to use in the computer simulations. The data were fitted to lognormal distributions. In addition, a model microstructure was produced by Dymet Ltd in which the grain structure was more widely distributed (Code F12) for comparison with the conventional material.

Micrographs of D1 and F12 are shown in Fig 2.

A lognormal distribution $f[x]$ is defined as:

$$f[x] = \frac{1}{x\sigma\sqrt{2\pi}} \exp \left[-\frac{(\ln(x) - \ln(\mu))^2}{2\sigma^2} \right] \quad (10)$$

where x is the variable, σ is a measure of the distribution width and μ is a measure of the distribution mean.

The lognormal distribution parameters, μ and σ (the geometric mean and standard deviation) can be ascertained by plotting the measured linear intercept data on lognormal probability axes, with a $\log_{10}$ abscissa, Figure 3. The straight line is a linear regression fit for the data from sample D1. It can be seen that a lognormal distribution is a good approximation to the measured distribution of intercepts of WC grain sizes. If the measured area distribution data is plotted on lognormal probability axes then it can be seen that the area data are also well described by a lognormal distribution, Figure 3. The lognormal distribution parameters derived from the linear and area measurements were found to be 1.08 and 1.25 for μ and 0.27 and 0.20 for σ for the intercept and area measurements respectively. The arithmetic number mean is higher than the geometric mean and the amount by which it is higher is dependent on the distribution width, σ . The arithmetic number mean is probably the best value to recommend as an indicator of size, as this is the value most widely measured and quoted in research papers. It is not recommended at this stage that this 2-D mean intercept value is converted to a value equivalent to that from a 3-D structure.

3.2 COMPUTER SIMULATION

Two approaches based on the use of computer simulation were used to assess the effectiveness of the linear intercept or area measurement techniques and their relevance to three dimensional structures [11]:

3.2.1 Tessellation of Truncated Trigonal Prism Slices

In the first approach a transputer-based method of producing simulated images of hardmetal structures was developed [12]. This procedure used an assumed distribution combined with information about the three dimensional WC grain shape to produce images that were equivalent to simulated sections through the hardmetal structure. These were compared with and shown to be similar in appearance to those produced using standard metallographic preparation techniques. The structure of a typical hardmetal was assumed to be made up of many crystals of WC bonded in a cobalt matrix. WC crystals develop the shape of a trigonal prism with truncated corners (TTP) when allowed to grow unconstrained [8,9]. However, little is yet known about what is the correct aspect ratio for these crystals in 3-D structures, whether the aspect ratio has single or distributed values or how the 3-D shapes are modified by constraint in real multiparticle structures. Consequently, an assumed shape, shown in Figure 4, with the edge length of the TTP arbitrarily chosen to have the value 7 compared with a height value of 3. These values were based on observations of the 3-D shape of WC crystals growing on the internal surfaces of small closed pores. A truncation value of about 1/6 was also arbitrarily chosen, where a value of 6 corresponds to the trigonal prism edge length and 1/6 of this is the cut-off length for the truncation. These values approximately correspond to a code of 45-25 for an equivalent "polychop" shape (see section 3.2.2).

The computer simulation technique was based on the random slicing of, and subsequent tessellation of, slices taken from the truncated tetragonal prisms of WC. The assumption that this process represents a true random slicing of a sintered WC/Co structure cannot be justified mathematically. However, a simulation of 3-D structures of two phase materials is computationally difficult with current computing systems and very time consuming. Consequently the 2-D approach was examined as a first step in gaining confidence in simulation methods. Also the 2-D sections of tessellated images were very effective in demonstrating the effects of differences in distribution width and shape.

A large number of random TTP sections were generated to give a distribution of different shapes [12]. An example of some randomly generated sections derived from individual TTP

crystals are shown in Figure 4. Randomly oriented lines were then used within each TTP slice to produce a distribution of intercept line lengths.

A fundamental assumption was made that packing TTPs in three dimensional space and then slicing the agglomeration was equivalent to slicing one TTP at random and then producing a tessellation of about 500 sections in two dimensional space. The sections were then picked at random from the pool of 500 precalculated polygonal TTP sections. A lognormal number distribution of 3-D TTPs was assumed so the sizes of the sections to be placed were calculated by scaling the vertices of the chosen polygonal TTP section by a lognormal probability density function with appropriate mean and variance. The sections were then sorted by area and a number of the largest sections were placed in order of decreasing size as predetermined seeds. The seeds were placed at random positions in a two dimensional box with random orientation. Each new section was placed so that it touched a previously placed section along one edge, which mimics features observed in the microstructures of real materials. This process continued until all the TTP sections were placed. The successfully placed particles typically accounted for 80-85% of the total area of the image which is similar to a micrograph of a WC/10 wt%Co hardmetal. Software routines were written that allowed the sections to be coloured with random shades of grey and with light or dark boundaries in order to simulate optical or electron micrographs.

The simulated image from the TTP tessellation program shown in Figure 5 was produced using the linear intercept lognormal distribution parameters obtained from measurements on hardmetal D1. The simulation technique is also very convenient for examining a number of additional input parameters such as the shape and size distribution of the TTPs. Figure 5 shows the effect of using TTP grains of all the same size (uniform) compared with the lognormally distributed sizes.

Although the lognormal distribution of three dimensional particle sizes used for the simulation produced realistic images, it was found that this resulted in an intercept length distribution which was dissimilar to the experimentally measured intercept length distribution [13]. There was a notable deviation at small intercept lengths. Far more small value intercepts (by number) were obtained from the computer simulated distributions. This effect has also been noted in measurement studies of TiB₂ cermet microstructures [13]. This may be because the real distribution of three dimensional particle sizes is not lognormal. Alternatively, rounding of the edges of the TTP and particle/particle contiguity effectively apply a cut-off in the intercept distribution [15,16]. There is also the possibility that real structures contain a collection of different TTP crystal shapes, rather than just one shape. This would clearly affect the final intercept distribution. Finally, the preparation and analysis of microstructures can round grain edges and blur them such that small intercepts are not counted.

3.2.2 Polychop

In the second approach a different computer program was written (called Polychop, a copy of which can be obtained from NPL) that will run on a standard PC. The program also contains shapes other than TTPs, for example a tetrakaidecahedron, a cube, a cylinder and a tetrahedron. The parameters that define the TTP can be varied to produce a wide range of shapes. For example at each extreme, flat plates or rod-like shapes can be constructed (Fig 6). These shapes were also used to explore the consequent distribution of 2-D intercepts and areas that can be generated from a 3-D shape. The program generates statistical data on each shape for a specified number of sections. The data includes information on areas, intercept lengths, vertices and numbers of sides. The program can be run with single sizes of shapes or as normal (a Gaussian) or lognormal distributions. The data are generated by randomly sectioning the shape (distribution).

Intercept data were also obtained from sections of the tetrakaidecahedron shape (Fig 7).

The effect of different TTP shapes was investigated by generating intercept data from random sections through four shapes coded 50-50; 50-25; 100-25; 20-25. These shapes are shown in Fig 6. The first number corresponds to the relative height to base ratio of the TTP with 100 equivalent to an elongated TTP and 25 corresponding to a thin plate. The second number equates to the amount of truncation, as can be seen in the diagram.

It was found that the best way of representing intercept distributions from the different shapes was to plot estimated area probabilities (from squaring the intercepts) as a size fraction against a normalised value of the intercept. This allowed populations of different size to be more easily compared. Fig 8 shows a corresponding plot for distributions from the 50-25 (equiaxed), 100-25 (rod like) and 20-25 (plate like) shapes compared with data from the M15 hardmetal material. The data were obtained from lognormally distributed populations of the TTPs with a distribution width (sigma) of 0.3. Fig 9 shows the distribution data from the hardmetal M15 compared with intercept data from the tetrakaidecahedron and TTP model shapes with a distribution width, σ , of 0.3. It is clearly not possible to differentiate between the TTP and the tetrakaidecahedron. Therefore Fig 10 shows the M15 hardmetal data plotted against data from the tetrakaidecahedron with different values of sigma. These plots indicate that a shape equivalent to the 50-25 TTP or the tetrakaidecahedron lognormally distributed in 3-D with a sigma of 0.3 gives the closest fit to measurements on conventional structures. Fig 10 also contains the data from hardmetal F12 which indicates that it is considerably more widely distributed than the conventional hardmetal M15. Also, Figs 8 and 10 indicate that it is quite difficult, if not impossible, to deconvolute the effects of distribution width and shape since a plate-like TTP gives equivalent area probabilities to those of a widely distributed equiaxed TTP. The plots in Fig 10 are very useful in answering the question "how uniform is a particular distribution". The data shown are for sigma values (measure of distribution width) of 0.01, 0.3 and 0.6. Clearly the nearer is a measured distribution to that corresponding to a distribution with a sigma of 0.01 then the more uniform is that structure.

3.3 AREA ESTIMATION

Direct measurement of the areas of WC grains is very time consuming, especially as up to 200 grains probably need to be measured to obtain reasonable distribution data. However, it is possible to use the intercept measurements, by squaring the individual values, to obtain an estimate of area. This is not obviously reasonable because the probabilities of measurement associated with small intercepts through the edges of large grains and the probabilities of measurement related to the selection of small or large grains are not easy to quantify. Consequently, the tetrakaidecahedron model shape was used to directly compare data obtained by squaring intercepts and by direct area measurement. This data was obtained for three distributions.

- single size tetrakaidecahedron
- lognormally distributed, $\sigma = 0.3$
- lognormally distributed, $\sigma = 0.6$

The results are shown in Fig 11 for size probabilities plotted against areas or estimated areas (intercept squared). The abscissa is plotted both directly and by normalising with the number average. It can be seen that for direct measurements the agreement between areas and estimated areas is very consistent. For the normalised plot there is a shift to higher values for the estimated area data but the trends (i.e. dependence on distribution width) are very similar. Clearly Fig 11 supports the use of intercept squared values as a measure of estimated area for investigating trends in distribution width.

3.4 SHORT INTERCEPTS

It is likely that in measurements on real microstructures that short intercept values are undercounted due to difficulties in image resolution and grain rounding. For this reason measurements from the structure of hardmetal M15 were compared with data from modelling a lognormally distributed (3-D) tetrakaidecahedron. The results are shown in Fig 12 plotted both as a number and a size probability. It can be seen that the number probability at low intercept values is much higher in the data from the model tetrakaidecahedron, as predicted. However, when the data are plotted as size probabilities this difference disappears, because clearly the fraction of area occupied by the small intercepts is tiny.

3.5 SERIAL SECTIONS

An 11 wt%Co/coarse grained WC hardmetal was used for the 3-D reconstruction measurements. A polished and etched section was prepared and marked with a diamond hardness indentation to act as an indicator and for calculating material removal rates during serial polishing. An image was obtained of the structure close to the indentation using a high resolution field emission scanning electron micrograph (FESEM). The sample was then repolished sequentially, each time removing a fraction of a micrometre of hardmetal, and reimaged. The amount removed, however, proved difficult to control. Fig 13 shows sequential FESEM images. The amounts removed between the sections varied between about 0.3 and 1 μm .

In order to reconstruct the 3-D structure of selected grains the coordinates of the vertices of the selected grains were identified in each serial section and these were used together with the slice thickness calculated from the dimensions of the diamond hardness indentation at each sectioning position. The slices were then combined using a computer work station and dedicated software to give the images shown in Fig 14.

It can be seen that the slice registration is reasonable, although not perfect, due to the difficulty of matching images from adjacent slices. The reconstructed grains were then used to generate a set of random intercept data for comparison with 2-D intercept distributions from the computer simulated distributions.

Intercept distributions, and thus estimated area probabilities, were obtained from random sections of the 3-D reconstructed shape in order to examine whether the 3-D reconstructed shapes resulted in intercept distributions similar to those from the model shapes. The model data was obtained from grains of all the same size, since not enough 3-D reconstructed grains were produced to allow distribution data to be obtained. The shape in Fig 14 was used for the 3-D reconstructed intercept data.

The results are shown in Fig 15. Also included are estimated area probabilities for random intercepts through a tetrakaidecahedron. The distribution for the 3-D reconstructed shape does not simply fit any of these distributions, although it tends towards that of the more equiaxed structures.

4 CONCLUSIONS

Computer simulation techniques have been developed for investigating microstructural variables in hardmetal microstructures based on observations of real hardmetal micrographs. The techniques can be used as tools for the interpretation of grain size distribution measurement data and to study the consequences of changes in the three dimensional size distribution on the two dimensional measurements. The technique also has potential as a source of reference images for calibrating standard measurement methods. The images were developed based on the assumption that 2-D tessellations were realistic in modelling distributions of 3-D sections.

The study resulted in the following conclusions

- The reconstruction of 3-D shapes from 2-D serial sections is difficult because of inaccuracies in measuring the thickness of each slice.
- The reconstructed shapes do not correspond to that of a Truncated Trigonal Prism (TTP), indicating that in real structures individual grains are constrained from attaining an equilibrium shape by adjacent grains.
- The use of a tetrakaidecahedron shape was reasonably equivalent to the use of an equiaxed TTP for modelling a distribution of sizes in 3-D and converting this to size probabilities plotted against normalised sizes. The tetrakaidecahedron would thus appear to be a reasonable representation of grain shape for close grain packing in hardmetals.
- Plots of intercept data were most discriminating between difference structures when area (estimated from squaring the intercepts) probabilities were plotted against normalised value of linear intercept.
- It is difficult to ascertain from intercept distribution plots whether the deviation from that corresponding to a single sized structure is due to differences in shape or differences in distribution width.
- Uncertainties associated with measurements of small intercept lengths have a marked effect on the number probability distributions but less so when the data are plotted as size probabilities (ie area or estimated area fractions).

Finally, it is unlikely that information on 3-D shapes can be directly obtained from typical intercept distribution data. More work is needed to evaluate additional methods of characterisation, such as shape factors (and their distribution).

5 RECOMMENDATIONS FOR GRAIN SIZE DISTRIBUTION MEASUREMENT

It is recommended that the following procedure is adopted for measuring the size distribution of WC grains:

- Prepare a suitable 2-D polished and etched section (see ref 4)
- Measure at least 200 WC intercept lengths in μm
- Square the intercept data to get an estimate of area
- Calculate cumulative size probabilities, by dividing each (estimated) area increment by the total (estimated) area
- Calculate an arithmetic mean linear intercept in μm (number average of intercept lengths)

NB: This value should be quoted as the mean grain size, while recognising that it was obtained from measurements on a 2-D plane. It should not be converted to a 3-D size.

- Normalise the intercept values by dividing by the arithmetic mean intercept

There are now two ways to plot the data:

- (a) for direct measurements of size distribution plotted against size.

In this case:

- Plot the cumulative size probability (estimated area) against estimated area (intercept squared).

- (b) for comparing the distribution widths of different materials.

In this case:

- Plot the cumulative size probability (estimated area) against the normalised linear intercept

Fig 16 can be used as a reference plot to estimate how wide is the distribution. Fig 16 gives equivalent data from a tetrakaidecahedron shape with lognormal distributions of $\sigma = 0.01, 0.3$ and 0.6 . A σ value of 0.01 is equivalent to a structure with all grains the same 3-D size.

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7 ACKNOWLEDGEMENTS

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FIGURE CAPTIONS

- Fig 1 Different types of WC size distribution plots using intercept data from two commercial hardmetals with a different WC grain size distributions
- Fig 2 Micrographs of conventional WC/Co (D1) and wide grain size distribution WC/Co (F12)
- Fig 3 Typical intercept and area distributions of WC in hardmetal D1 (6% Co)
- Fig 4 Truncated trigonal prism, TTP, and randomly generated sections
- Fig 5 Simulated FESEM image of WC/Co with lognormal and uniform WC size distributions
- Fig 6 Truncated trigonal prisms; flat plate, equiaxed and needle shapes
- Fig 7 Polychop model of tetrakaidecahedron
- Fig 8 Effect of WC shape on distribution plots
- Fig 9 Comparison of WC size distribution with TTP and tetrakaidecahedron models
- Fig 10 Effect of WC size distribution on distribution plots
- Fig 11 Comparison of size probabilities for areas directly measured or estimated by squaring intercepts
- Fig 12 Effects of counting short intercepts on number and size probabilities
- Fig 13 WC/Co serial sectioned SEM images, numbered from 11C13-11C18
- Fig 14 Reconstructed WC grains from serial sections
- Fig 15 WC size distribution from 3-D reconstructed shape compared with polychop TTP and tetrakaidecahedron
- Fig 16 Reference 2-D size probability plots for 3-D tetrakaidecahedron distributions with $\sigma = 0.01, 0.3$ and 0.6

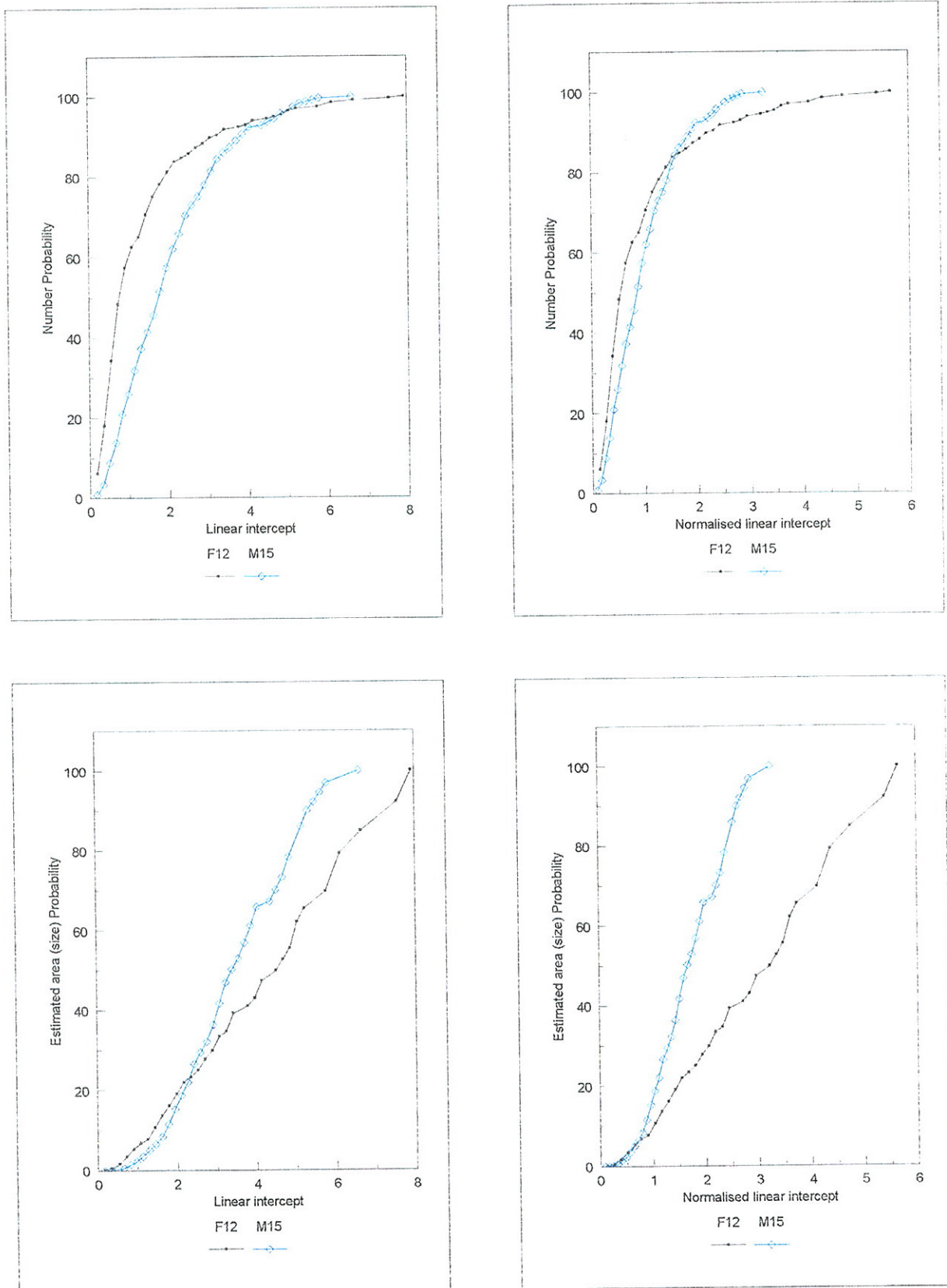


Fig 1 Different types of WC size distribution plots using intercept data from two commercial hardmetals with a different WC grain size distributions



Fig 2 Micrographs of conventional WC/Co (D1) and wide grain size distribution WC/Co (F12)

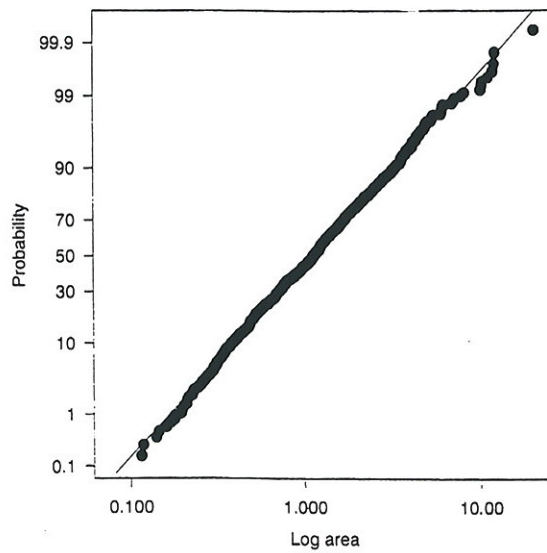
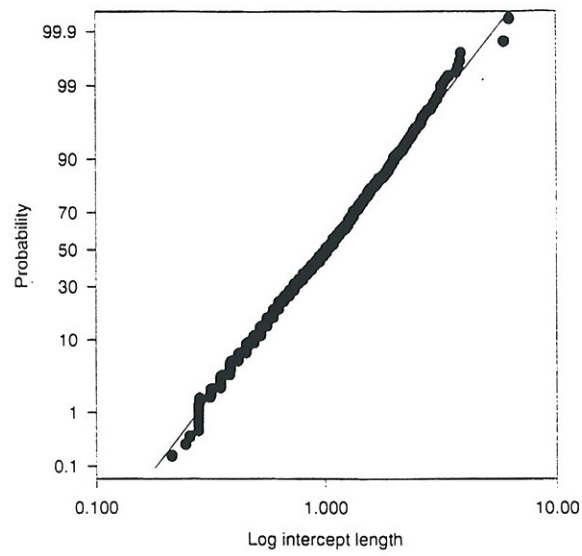
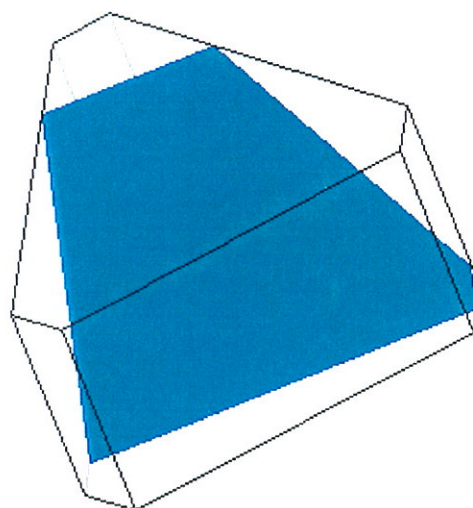


Fig 3 Typical intercept and area distributions of WC in hardmetal D1 (6% Co)

Truncated Trigonal Prism *Random section*

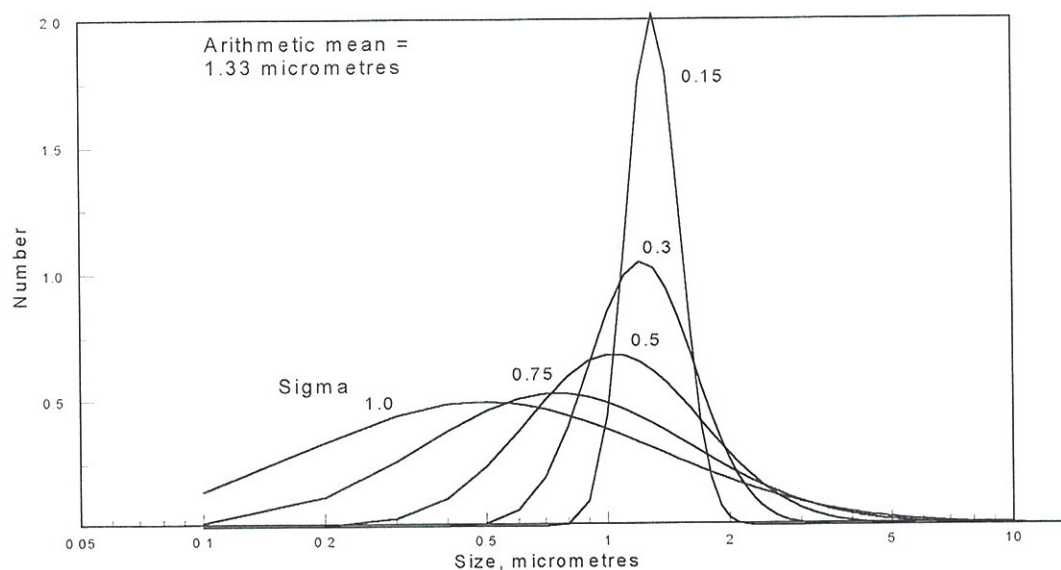
Sides=5 , Area=0.404807



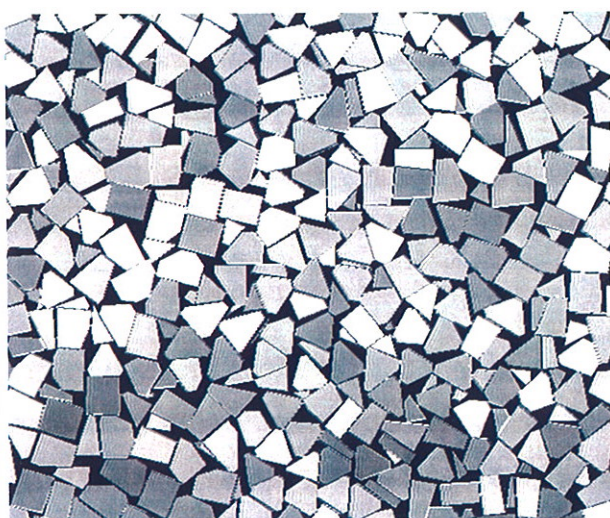
Length=1

Fig 4 Truncated trigonal prism, TTP, and randomly generated sections

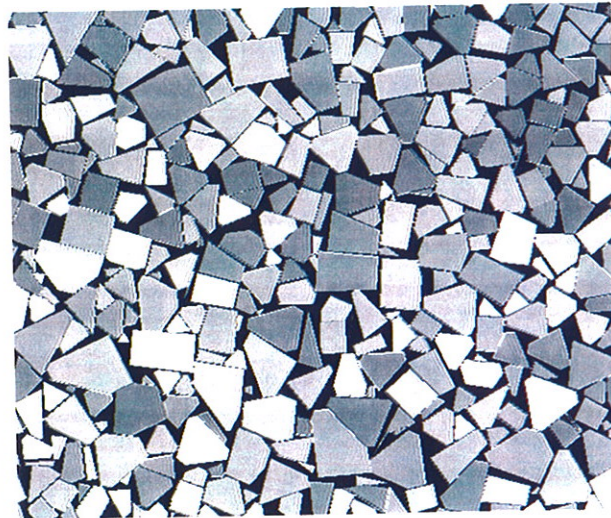
Grain size distribution characteristics



The plot shows number distribution histograms for the same arithmetic mean but different sigma (width) values.



Sigma = 0



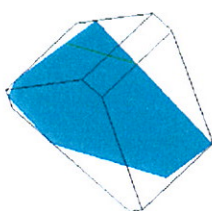
Sigma = 0.3

Computer simulated images of WC/Co structures with a sigma value of 0 (all grains same size) and a sigma value of 0.3 (typical of standard commercial materials).

Fig 5 Simulated FESEM image of WC/Co with lognormal and uniform WC size distributions

Truncated Trigonal Prism *Random sections*

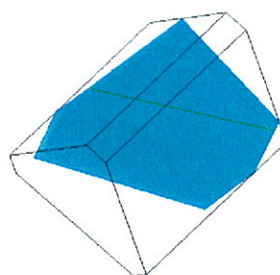
Sides=6, Area=4.20277, Intercept=1.162470 ± 0.000001, Scaling=2.397970



Length=2.397970

50-25

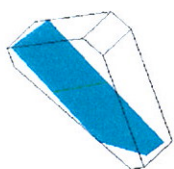
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Length=4.688673

100-25

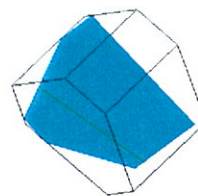
Sides=6, Area=2.357539, Intercept=1.008645 ± 0.000001, Scaling=3.815505



Length=3.815505

20-25

Sides=6, Area=1.233467, Intercept=1.132488 ± 0.000000, Scaling=2.025880



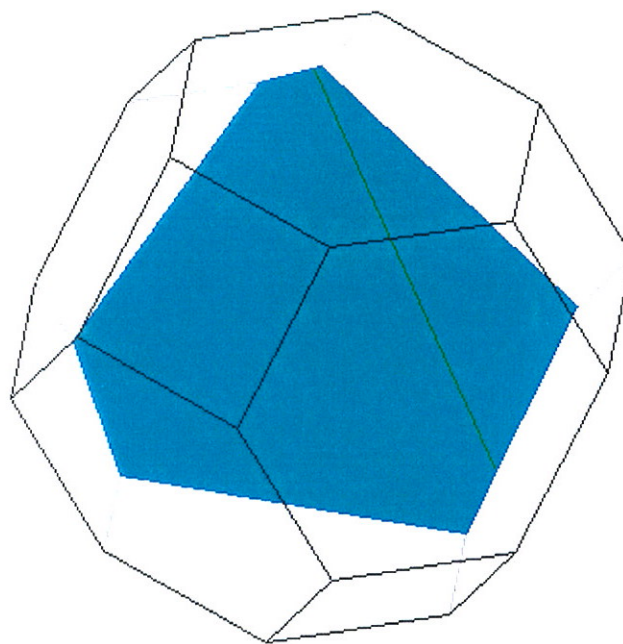
Length=2.025880

50-50

Fig 6 Truncated trigonal prisms; flat plate, equiaxed and needle shapes

TETRAKAIDECAHEDRON

Sides=6 , Area=25.109135, Intercept= 5.731701 ± 0.000004 , Scaling=2.629098



Length=2.629098

NPL 

Fig 7 Polychop model of tetrakaidecahedron

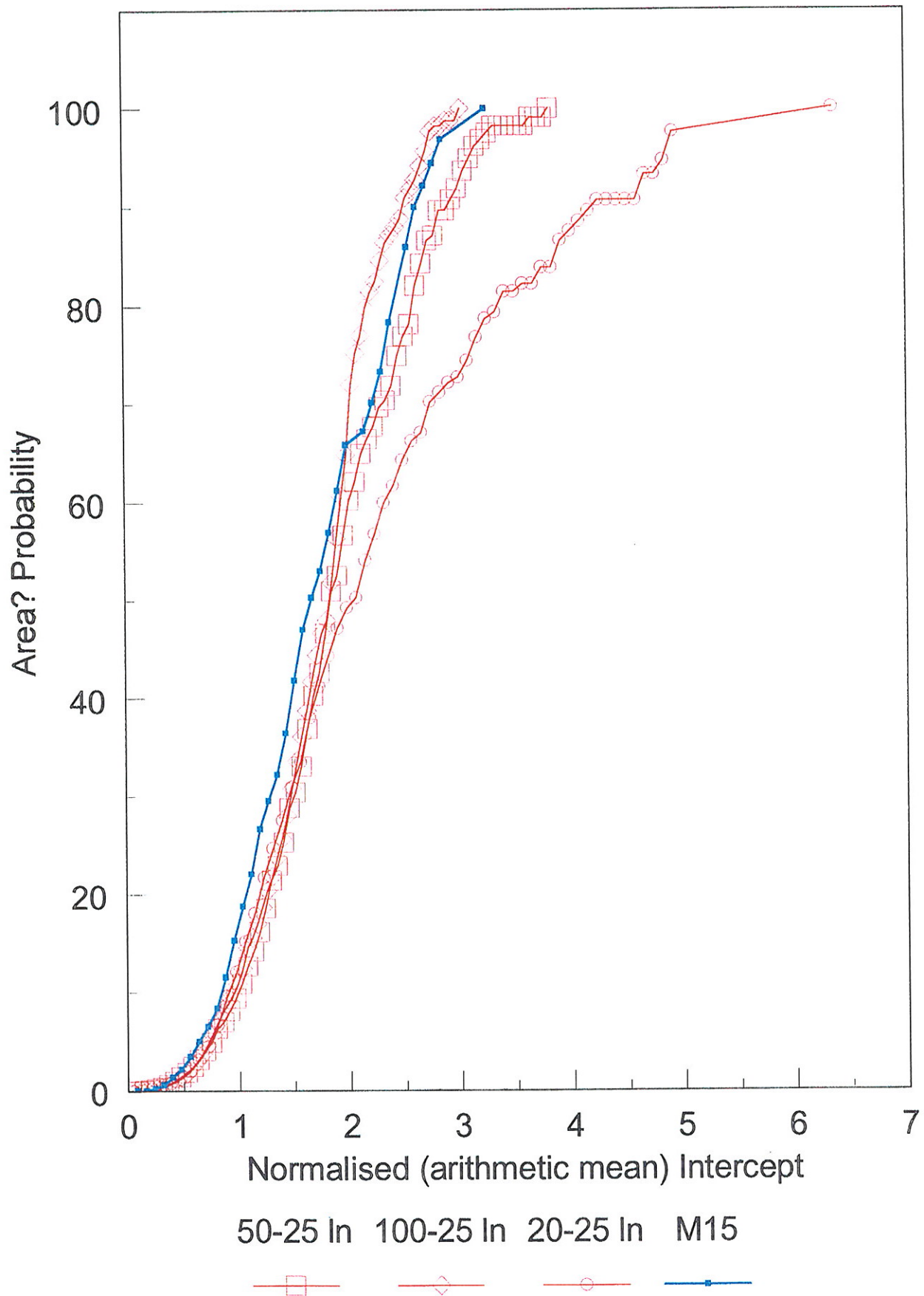


Fig 8 Effect of WC shape on distribution plots

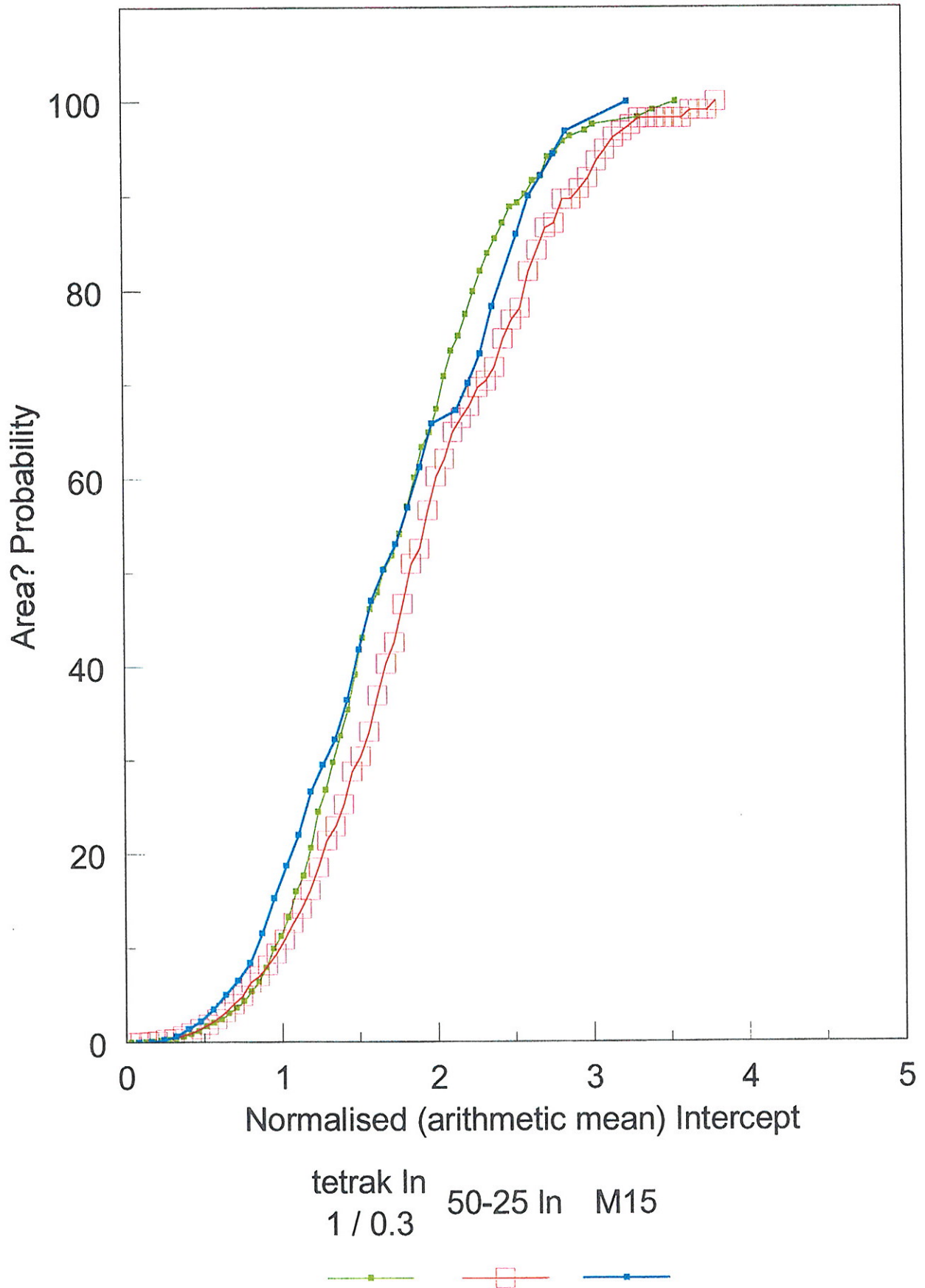


Fig 9 Comparison of WC size distribution with TTP and tetrakaidecahedron models

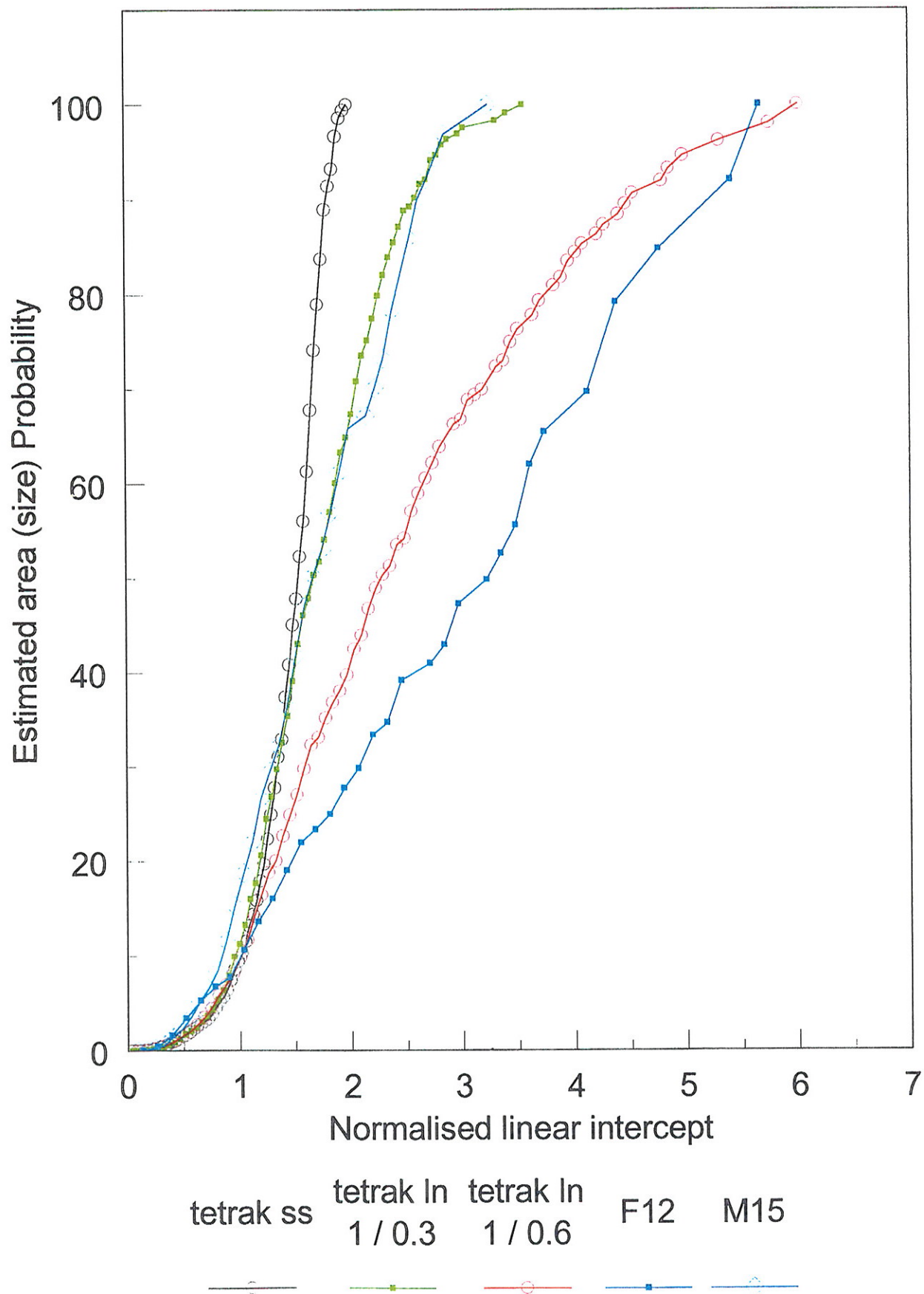


Fig 10 Effect of WC size distribution on distribution plots

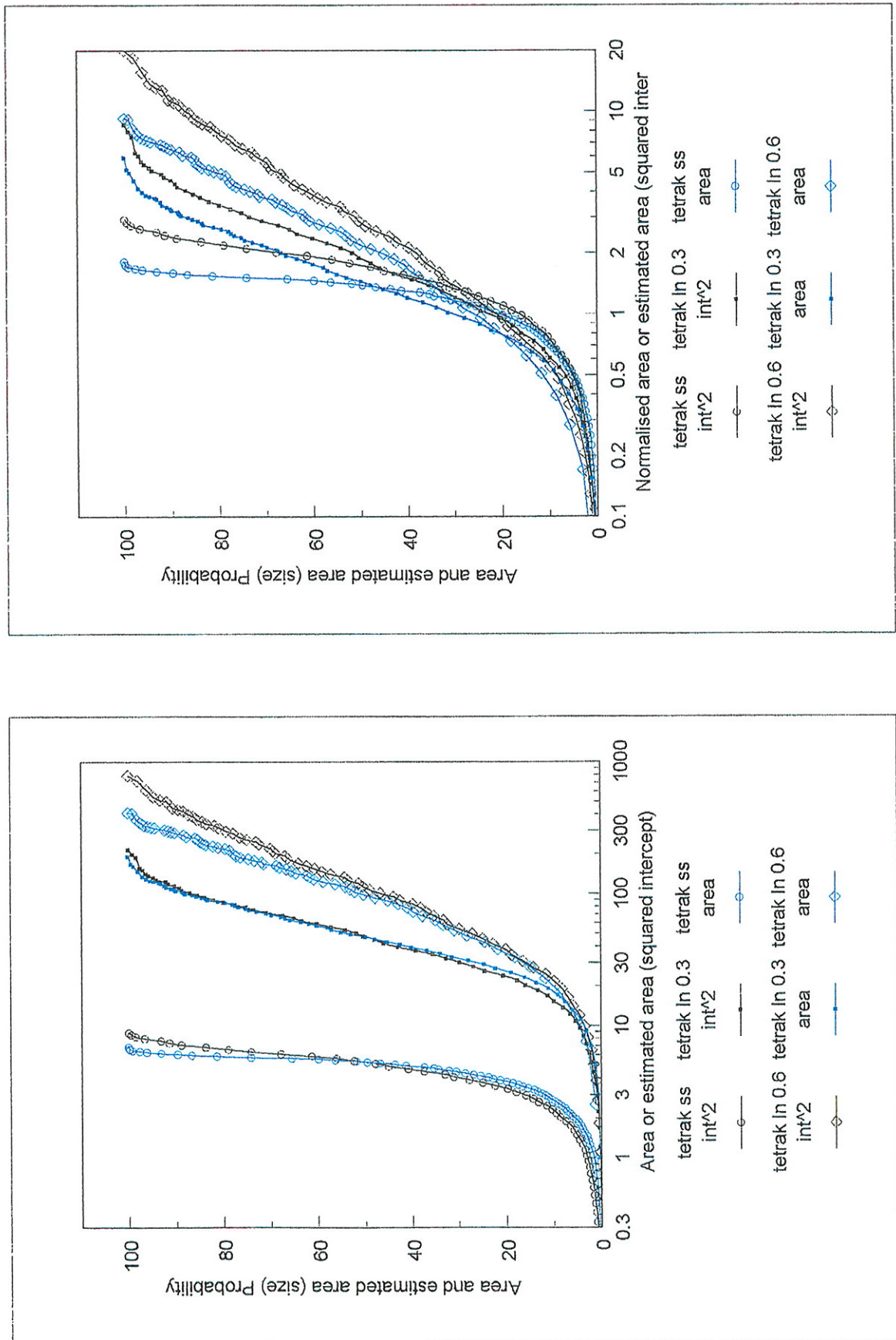


Fig 11 Comparison of size probabilities for areas directly measured or estimated by squaring intercepts

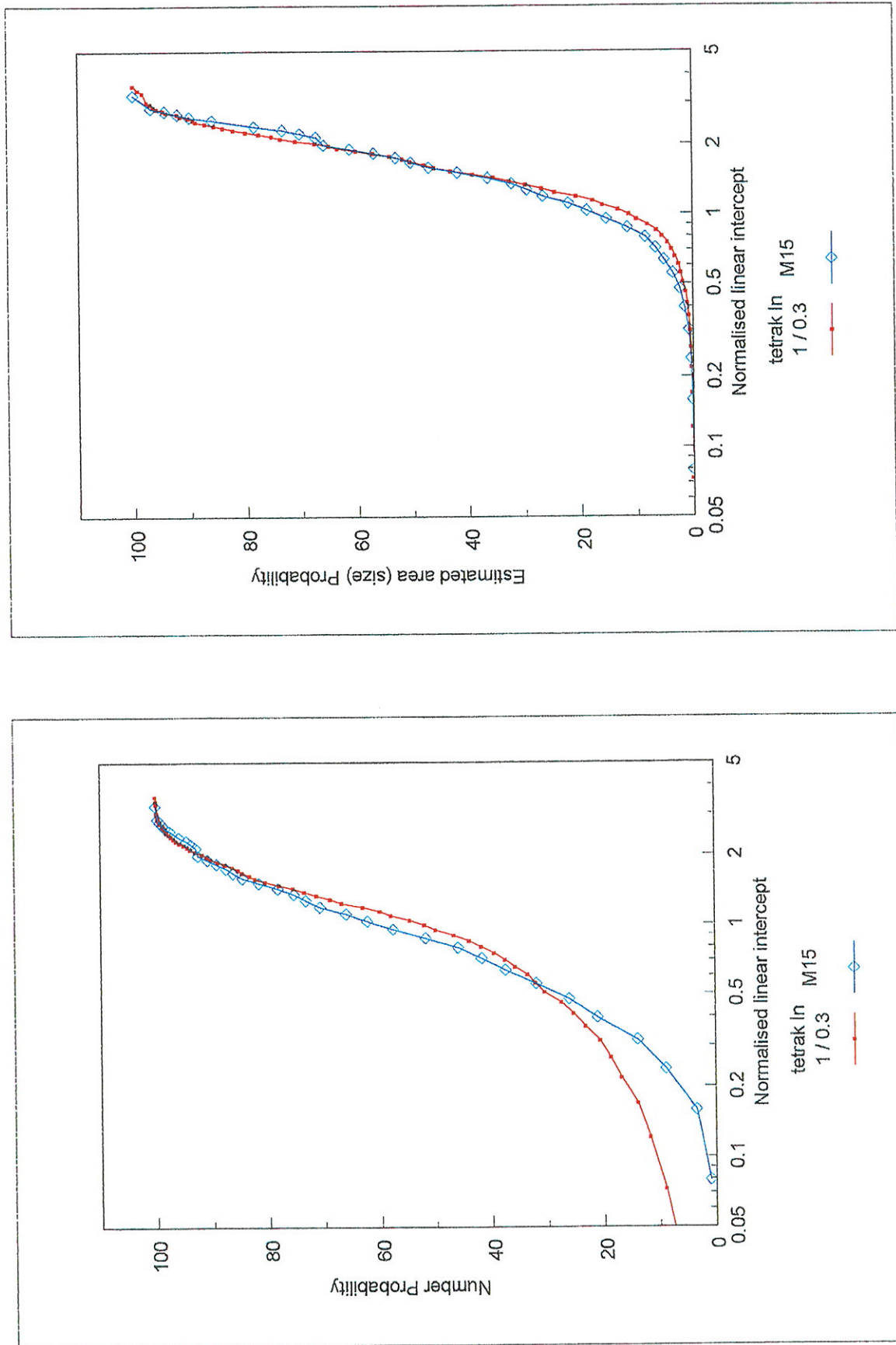


Fig 12 Effects of counting short intercepts on number and size probabilities

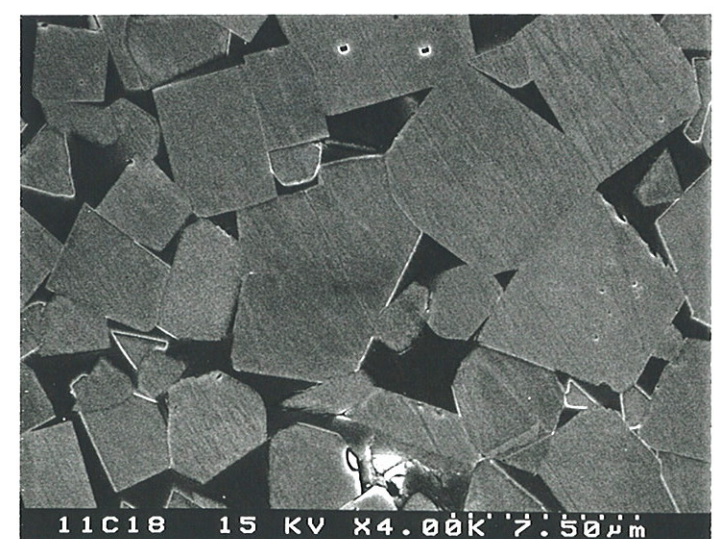
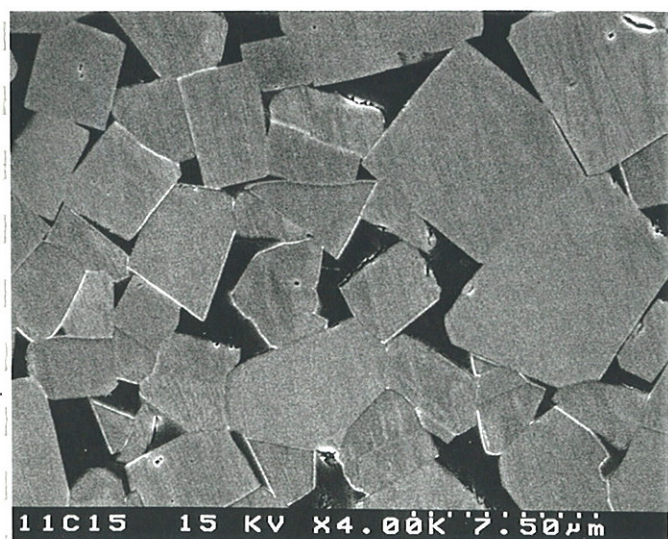
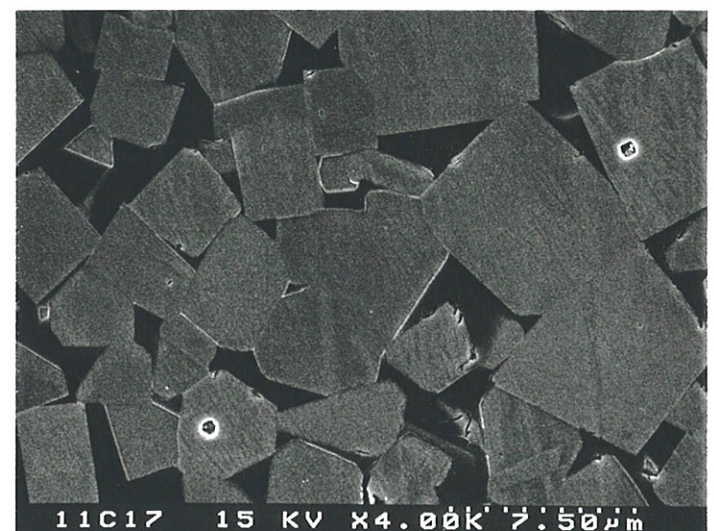
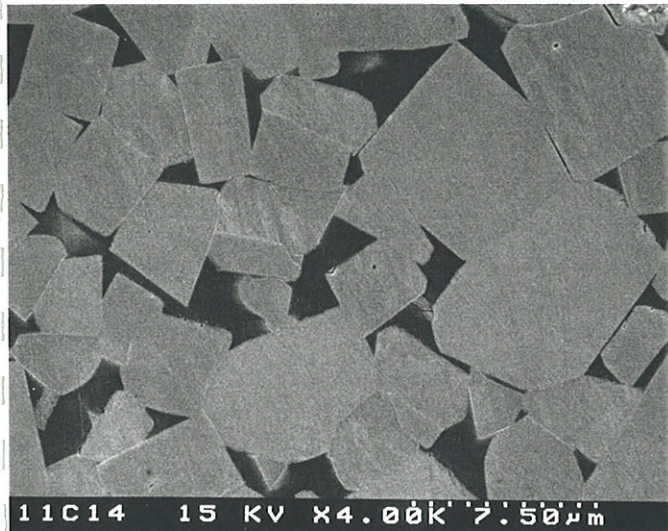
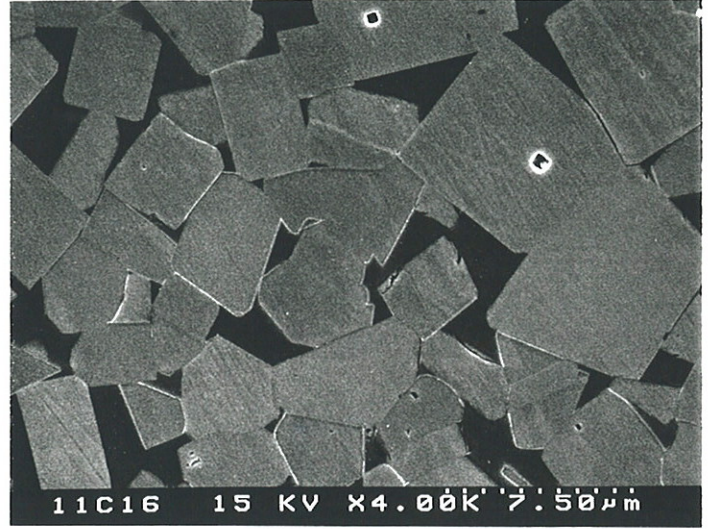
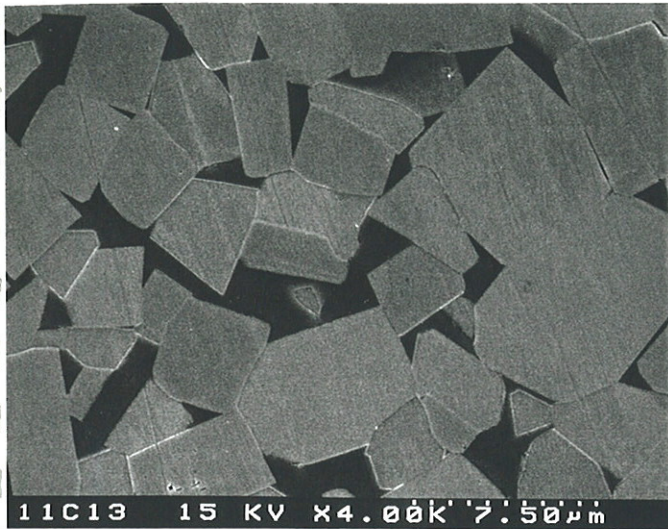


Fig 13 WC/Co serial sectioned SEM images, numbered from 11C13-11C18

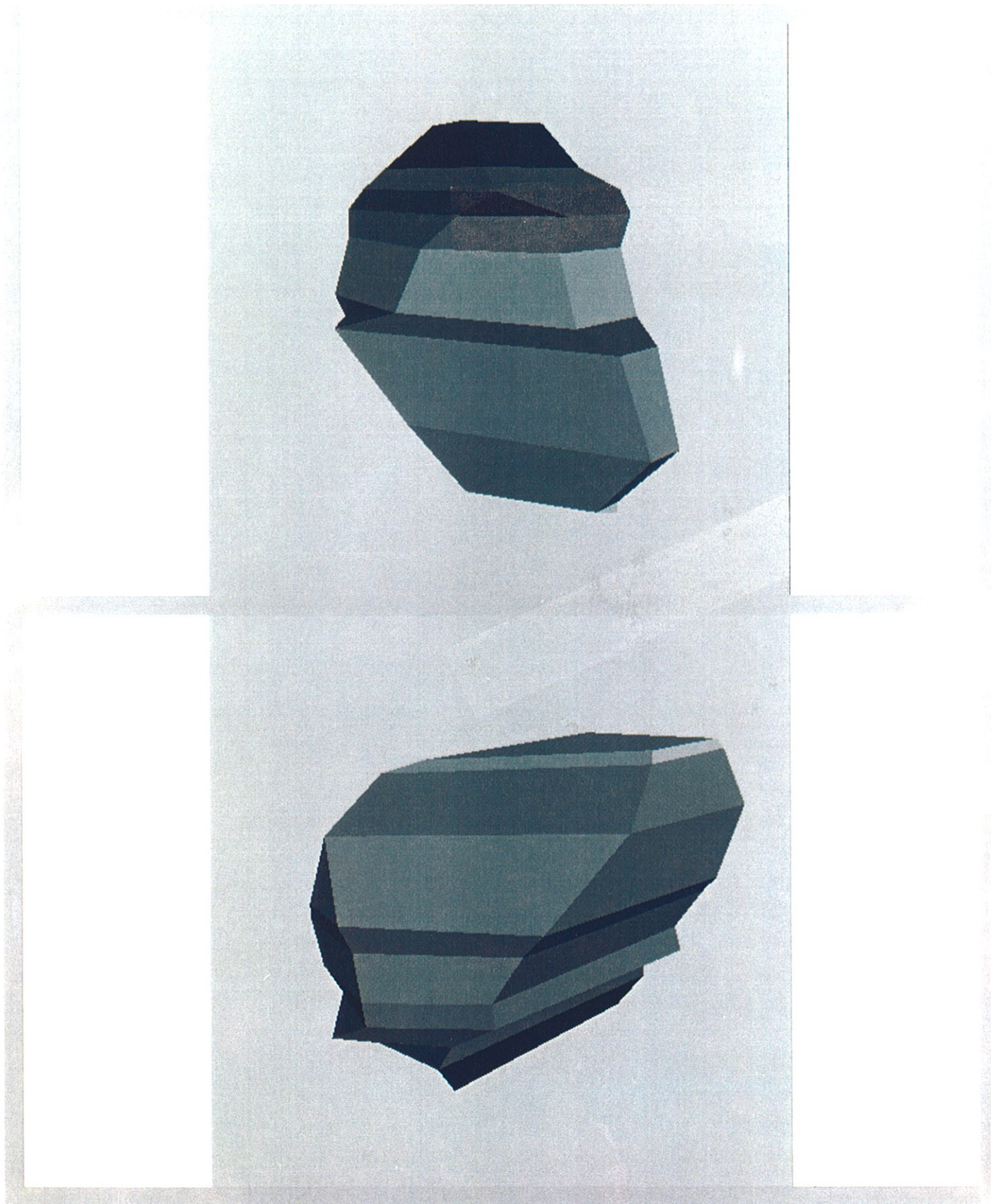


Fig 14 Reconstructed WC grains from serial sections

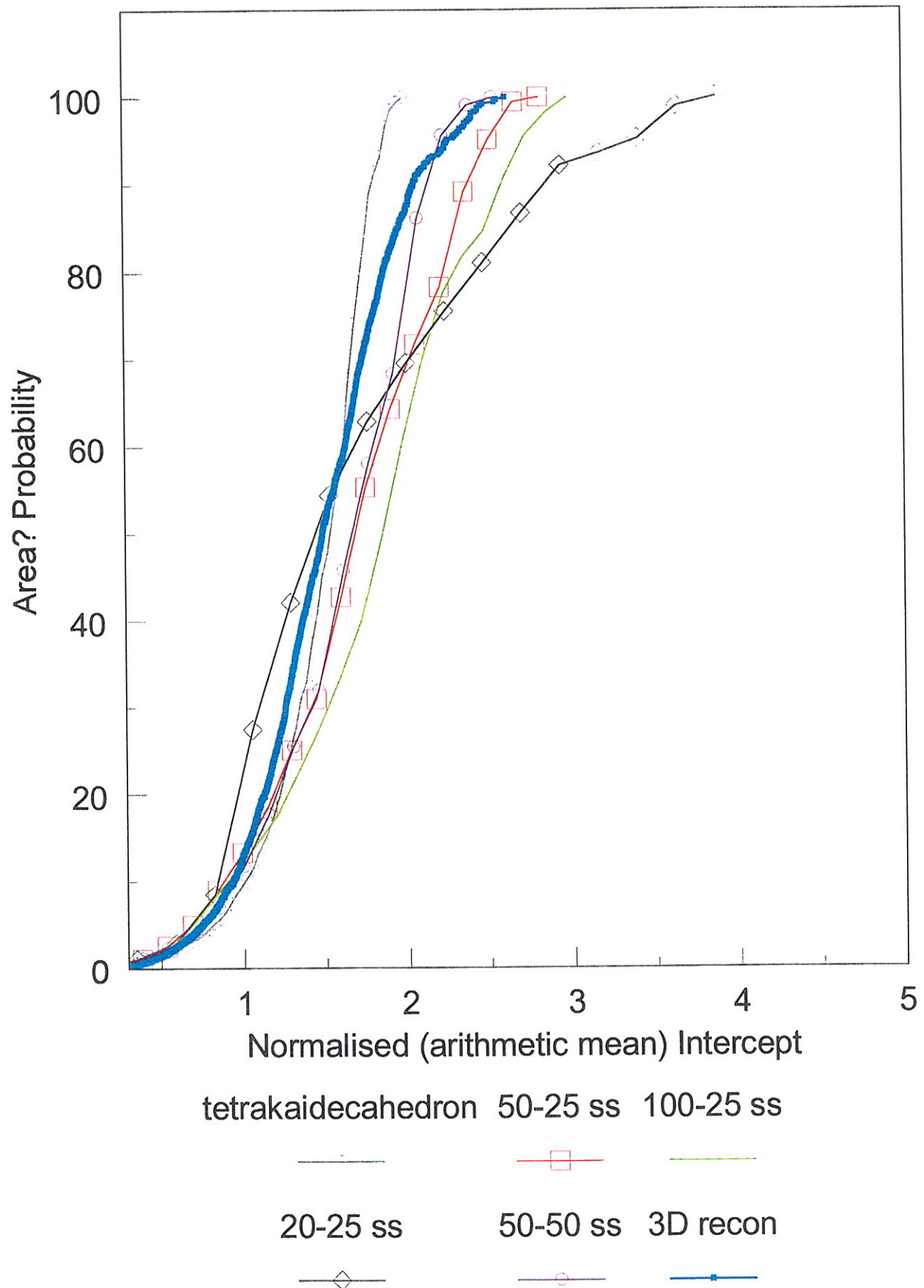


Fig 15 WC size distribution from 3-D reconstructed shape compared with polychop TTP and tetrakaidecahedron

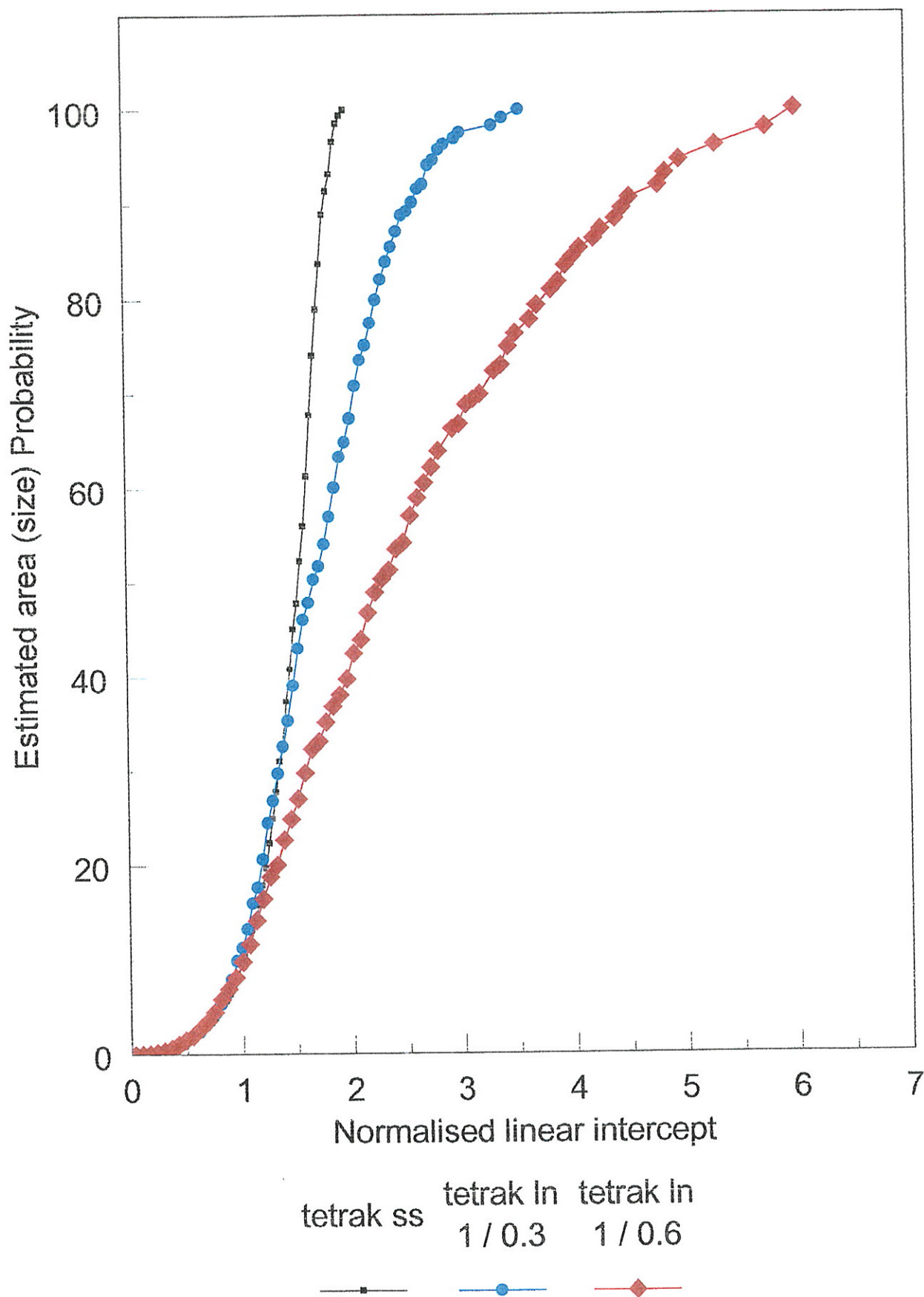


Fig 16 Reference 2-D size probability plots for 3-D tetrakaidecahedron distributions with $\sigma = 0.01, 0.3$ and 0.6