

**A Comparison of the
Linear Intercept and
Equivalent Circle
Methods for Grain Size
Measurement in WC/Co
Hardmetals**

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ABSTRACT

A computer model (POLYCHOP; developed at NPL) was used to calculate the equivalent circle diameter (ECD) and linear intercept (LI) grain size for a range of different crystal shapes, including a cube, a tetrakaidekahedron and a truncated trigonal prism. The effects of differences in size distribution and shape factors were studied and the results were compared with experimental measurements of the ECD and LI on sintered WC/Co hardmetals. Both the model and experimental results showed a linear correlation between ECD and LI for materials with grain sizes ranging from 0.5 to 5 μm . However, the model calculations, although showing the same trend, did not agree with the measurements. Possible reasons for this discrepancy, such as the difficulty of measurement of small grains, are discussed.

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1 INTRODUCTION

Crystal grain size is one of the most important factors in controlling the properties of materials. For example, the strength, toughness and hardness are all important engineering properties that are strongly influenced by this parameter. For this reason it is important to have standard methods for its measurement with commonly used and agreed terminology.

WC/Co hardmetals are manufactured from powders with the objective of producing hard and strong tool materials by controlling the grain (crystal) size of the WC phase. A typical micrograph of WC/Co is shown in Fig 1. The grey crystals are the WC phase. The black contrast regions between the WC grains is the cobalt-tungsten-carbon alloy binder phase.

There are currently two methods in use to estimate the grain size of WC/Co hardmetals [1-6] – one is based on the measurement of an equivalent circle diameter (ECD) and another on the use of linear intercepts (LI). Variability between the different methods impedes the comparability of data, and inhibits trade and new product development through the lack of a common set of results. This report investigates the relation between the two methods, both by modelling and experiment.

The LI method is based on the number average lengths of intercepts through each crystal/grain along a line drawn across the material surface. The ECD method is based on the number average area of the grain/crystals. The average area is converted to an equivalent circle diameter (ECD) as a measure of size. Both methods are justified, but find different values.



Fig 1 WC/Co structure.

The factors which may affect estimates of the grain size include the size and shape of the grains, their height to width ratios and the distribution of the size of the grains. In particular, determining the effect of the distribution of grain size in the material may be important. These factors can affect the two measurement methods in different ways, thus perhaps changing the relationship between the values found.

The grain size distribution data can be plotted as cumulative probability plots with the abscissa (x) on a linear or logarithmic scale. Historically, this has been the preferred method for plotting WC size distribution data, where a number probability is used for the ordinate and the size is plotted as log size on the abscissa. It has been found that linear intercept measurements plotted in this way have a lognormal distribution, giving a straight line when lognormal probability paper is used. The equation for lognormal probability is:

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

where σ is a measure of the distribution width and μ_g is the geometric mean. This does not plot conveniently mathematically, but many software packages have numerical approximations to the function, permitting σ to be determined. The use of probability paper generally gives a straight line for data for current commercial hardmetals, indicating that the parameter, σ , is the width of the lognormal distribution. Linear intercept measurements of WC grains have lognormal number distributions.

Modelling

The computer programme POLYCHOP was used to model the crystals and to calculate the average area and intercept from a sample containing a fixed number of crystals. POLYCHOP simulates the crystals in a material by taking intercept and cross-sectional area measurements of the crystal in different random orientations [7].

Three shapes were investigated; cubic, tetrakaidecahedral (TKC) and truncated triangular prism (TTP) crystals, Figs 2 and 3. Different size distributions of the crystals were used to simulate the variation of crystal size within a material. A lognormal distribution was chosen with σ values of 0.0, 0.1, 0.3 and 0.5, where σ is a measure of distribution width. The sizes of the crystal were also varied with scaling factors of 1, 2, 4 and 8, doubling the size of crystal each time. The effect of varying the number of grains counted was examined between about 100 and 100,000.

When comparing the modelled results with measured results the TTP shaped model was used as it is assumed to approximate most accurately to the shape of crystals in WC/Co materials. For the modelling exercise the dimensions of the TTP were edge, height and truncation 0.5. (Fig 3).

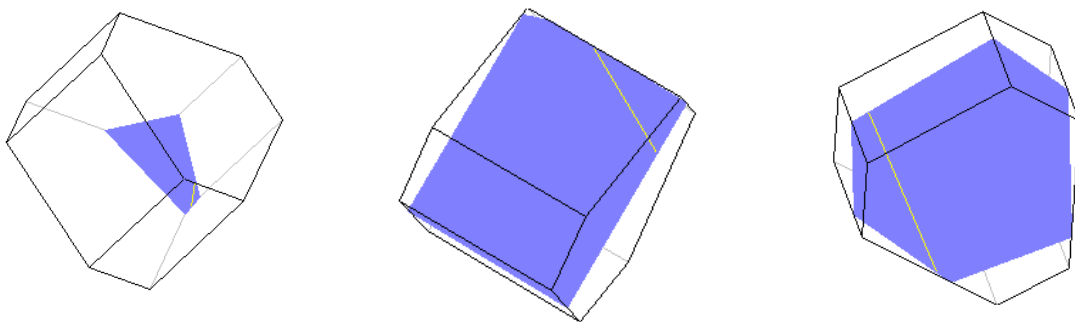
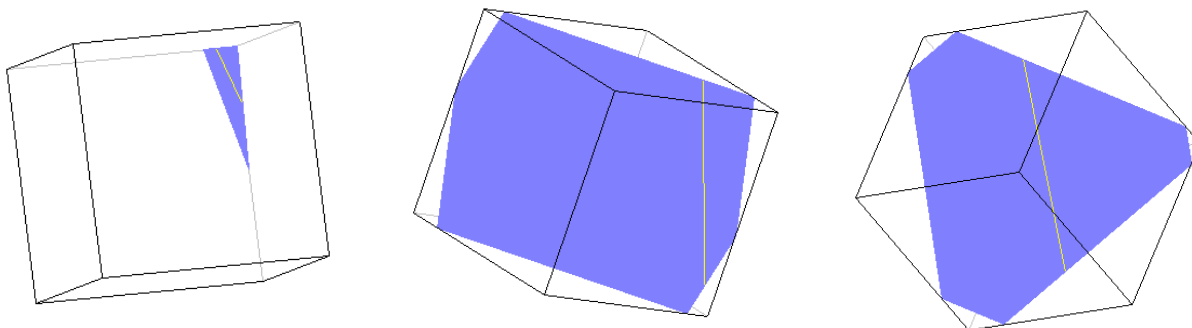
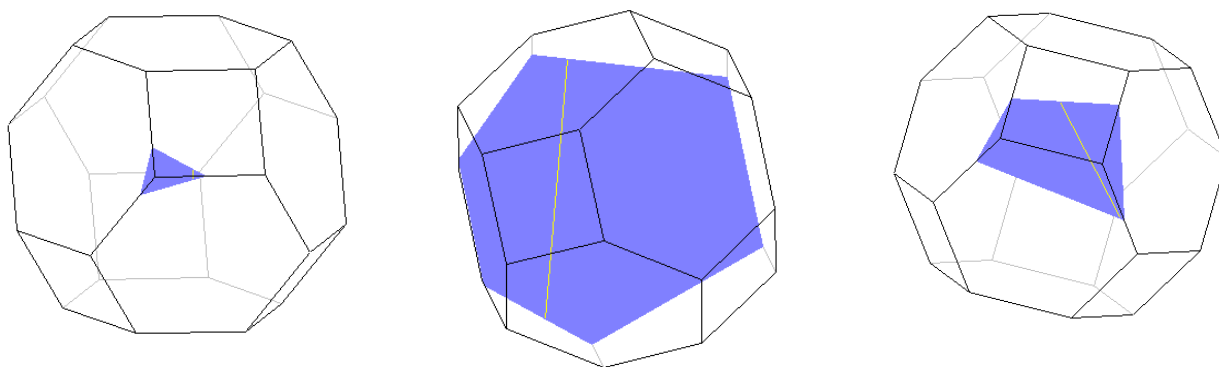
TTPCubeTKC

Fig 2 POLYCHOP shapes with typical random cross sections and intercept lengths.

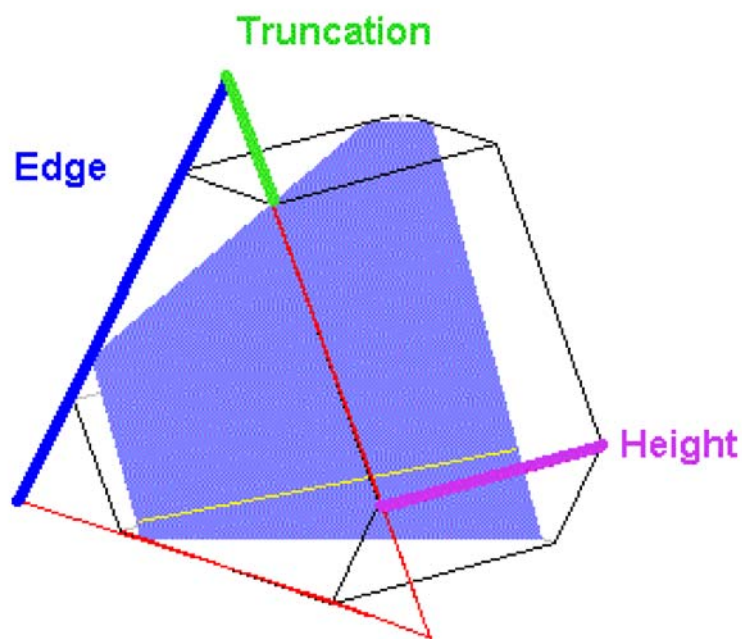


Fig 3 Diagram showing TTP measurements

2 MATERIALS AND EXPERIMENTS

Linear intercepts and equivalent circle diameter measurements were obtained on a range of WC/Co hardmetals (Table 1). All the materials investigated were manufactured in the two-phase, WC/Co region, and magnetic data are also given in Table 1. The coercivity data were supplied by Marshalls Hard Metals Ltd. The magnetic moments were measured at NPL using LDJ[®] equipment to support these observations.

Table 1
Hardmetal Properties

Grade	wt% Co	Volume Fraction WC	Coercivity kA m^{-1}	Magnetic moment $\mu\text{T m}^3 \text{ kg}^{-1}$	Grade	Wt% Co	Volume Fraction WC	Coercivity kA m^{-1}	Magnetic moment $\mu\text{T m}^3 \text{ kg}^{-1}$
T25	24.2	0.55	6.8	2.82	G10	9.90	0.82	6.5	1.48
D10	10.0	0.82	9.8	1.35	CW25C	24.44	0.54	4.4	3.52
T	9.1	0.83	8.4	1.10	CW20C	19.68	0.62	4.1	2.87
UC9	9.8	0.82	4.0	1.35	TC222	6.09	0.88	2.5	0.98
MA11	11.7	0.78	3.8	1.64	K3560	9.69	0.82	4.6	1.37
K3520	20.3	0.62	5.1	2.88	M15	15.2	0.7	5.1	2.11

The samples were polished and etched in Murakami's reagent and images for subsequent measurement of grain size were obtained either using an optical microscope at $\times 1600$ or in a scanning electron microscope at different magnifications.

One image of each sample was analysed to calculate the average ECD. The weight % Co was converted to a volume % Co(F) and then the area of the image was multiplied by a factor (1-F) to get the total area occupied by WC. The number of grains was counted. Grains that were on the edge of the image were counted as half grains. The average area of each grain was calculated and then the ECD calculated from this value.

3 RESULTS AND DISCUSSION

3.1 MEASUREMENTS

Samples of 11 hardmetals with a range of grain size were polished and imaged using a Scanning Electron Microscope (SEM). These images were then analysed by 2 different operators, (A) and (B) to calculate ECD values for each sample. The ECD values were then compared to the LI data taken from recent NPL reports [8,9]. These results show a clear linear relationship (Fig 4) between the two parameters. The individual results are given in Table 2.

Table 2
ECD/LI Measurements

Operator A			Operator B		
Grade	LI	ECD	Grade	LI	ECD
T25	0.77	1.10	T25	0.77	1.06
D10	1.05	1.27	D10	1.05	1.16
T	1.68	2.00	T	1.68	1.80
UC9	4.97	5.75	UC9	4.97	5.51
MA11	5.16	6.38	MA11	5.16	5.98
K3520	2.30	2.62	K3520	2.30	2.23
G10	1.76	2.66	G10	1.76	2.29
CW25C	2.14	2.73	CW25C	2.14	2.30
CW20C	2.17	2.50	CW20C	2.17	2.25
TC222	3.59	4.71	TC222	3.59	4.13
K3560	2.75	3.30	K3560	2.75	3.26
M15	2.04	2.60	M15	2.04	2.30

The results show good agreement between the relation between ECD and LI, Fig 4 illustrating this through the similarity of the slopes of the two lines. There is a clear linear relationship, but there is a systematic difference between the results of operators (A) and (B) where the ECD calculated by (A) is always bigger than that of (B). The mean value of the slope is 1.15. This correlation is in good agreement with previous work cited by De Hoff and Rhines in the book "Quantitative Microscopy" [10] where it is stated that an empirical relation had been found in measurements in aluminium and ferrous alloys where

$$LI = \sqrt{\bar{A}}$$

where $\bar{A}$ is the average grain area (Jefferies method). Thus since $\bar{A} = \frac{\pi}{4} (ECD)^2$

$$ECD = \sqrt{\frac{4}{\pi}} \sqrt{\bar{A}} = 1.13 \sqrt{\bar{A}} = 1.13 LI$$

Clearly the measurements on WC/Co alloys on the comparison between ECD and LI agree well with previously published work on other engineering alloys.

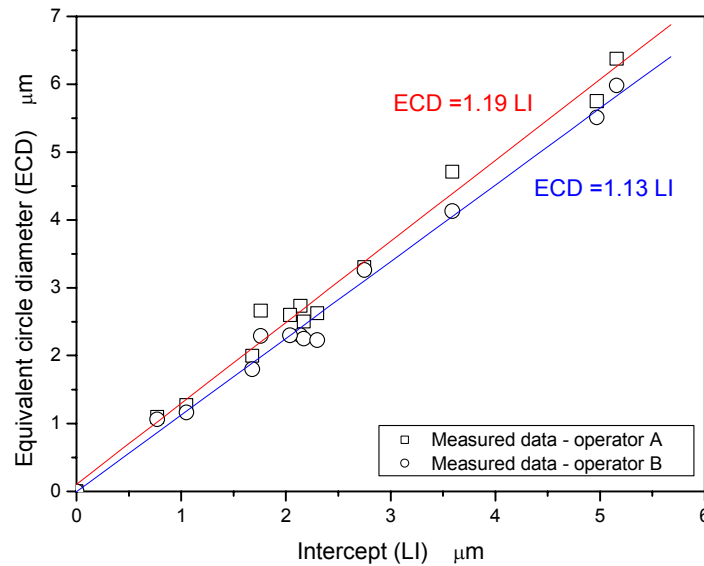


Fig 4 Measured ECD against LI data

3.2 MODELLING

3.2.1 Effects of Size, Distribution and Sample Number

Cumulative distribution graphs for computed linear intercept and area plotted against size are shown in Figs 5-8 and 9-12 respectively for each crystal shape, size and size distribution. The data are plotted on a linear scale for the y ordinate and a log scale for the x ordinate to enable uniform comparisons to be made. A set of parallel frequency distribution plots are shown in Figs 13-20. The sample size used for generating these graphs was N=1000.

The cumulative plots show similar trends for all the shapes. There was no effect of scaling factor, confirming internal consistency of the program and generating information for comparing ECD and LI averages. The frequency distributions show similar trends with the expected peaks corresponding to face edge lengths in the distributions for $\sigma=0$ (i.e. all crystals the same size). For higher values of σ these peaks systematically reduce, as expected.

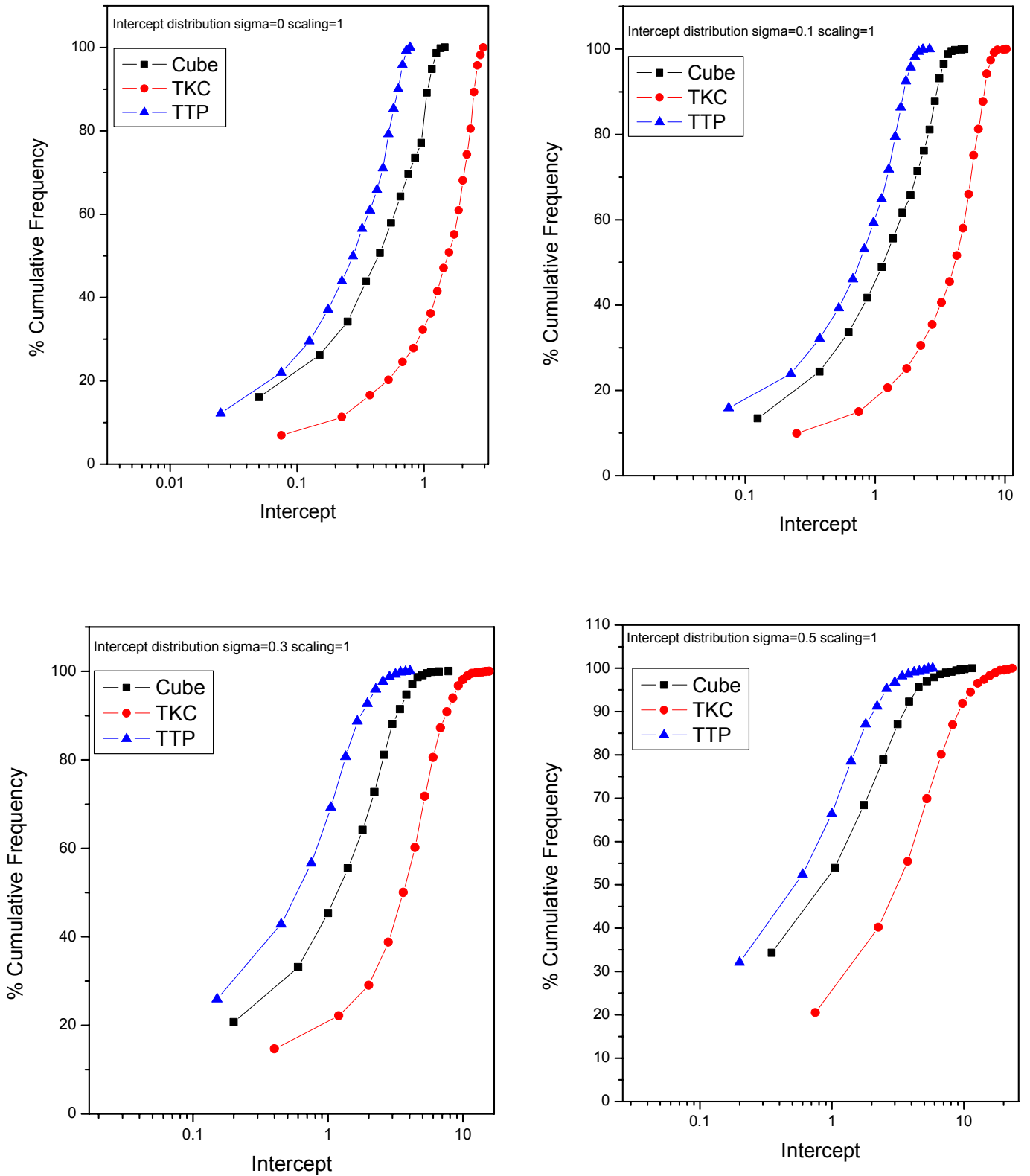


Fig 5 Computed cumulative number distribution plots for intercepts through each crystal shape showing variation with distribution width at a scaling factor of 1.

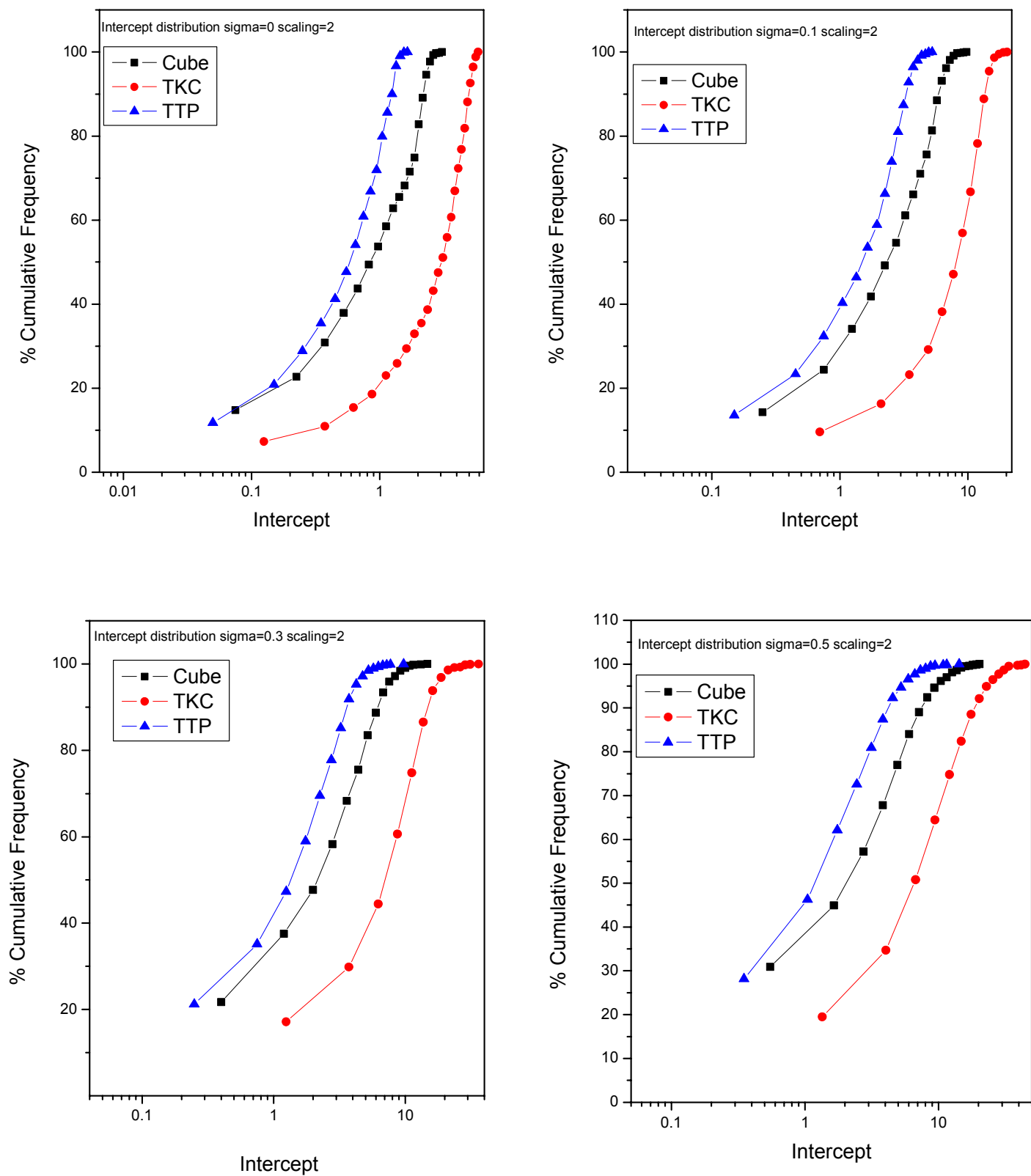


Fig 6 Computed cumulative number distribution plots for intercepts through each crystal shape showing variation with distribution width at a scaling factor of 2.

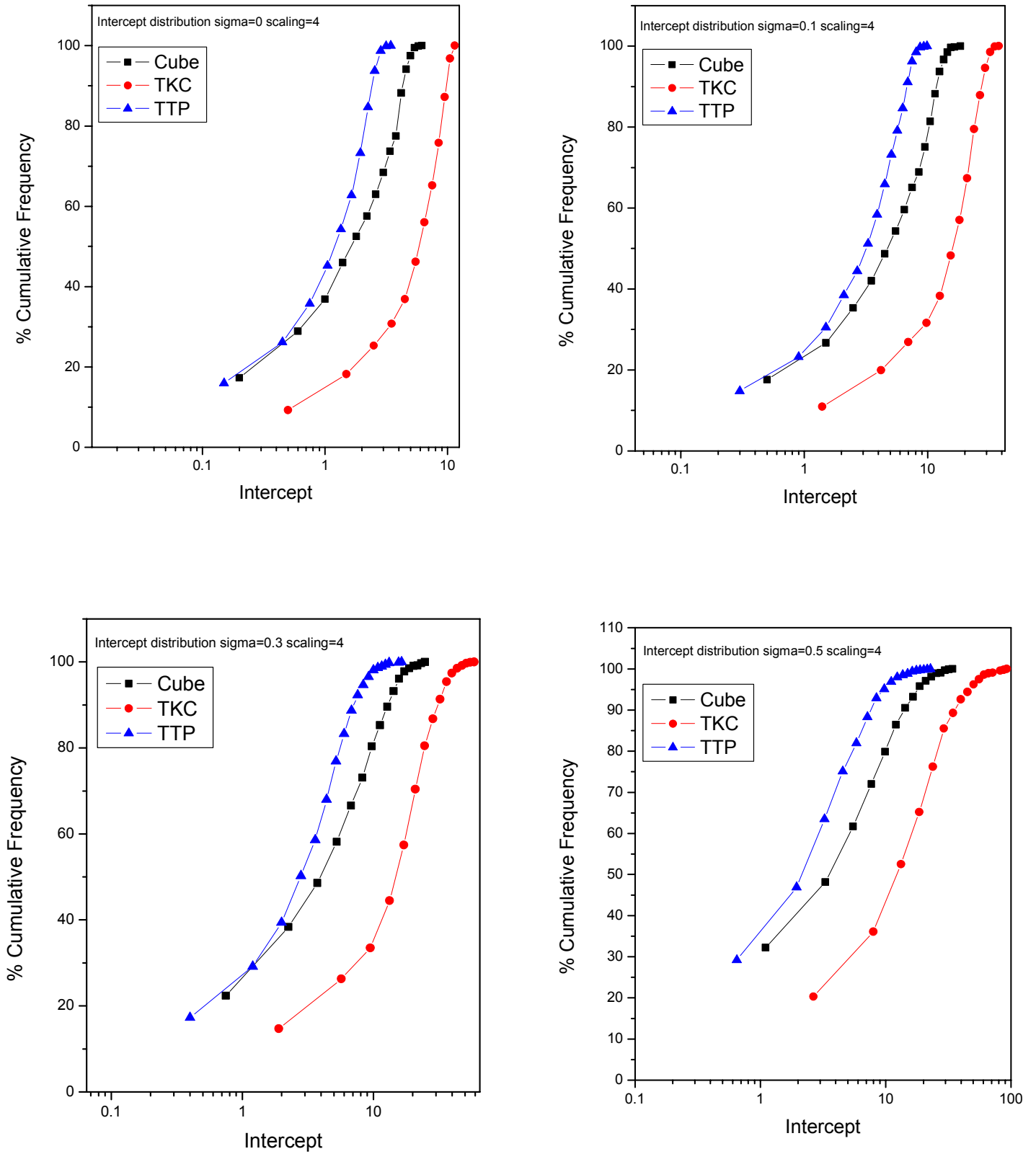


Fig 7 Computed cumulative number distribution plots for intercepts through each crystal shape showing variation with distribution width at a scaling factor of 4.

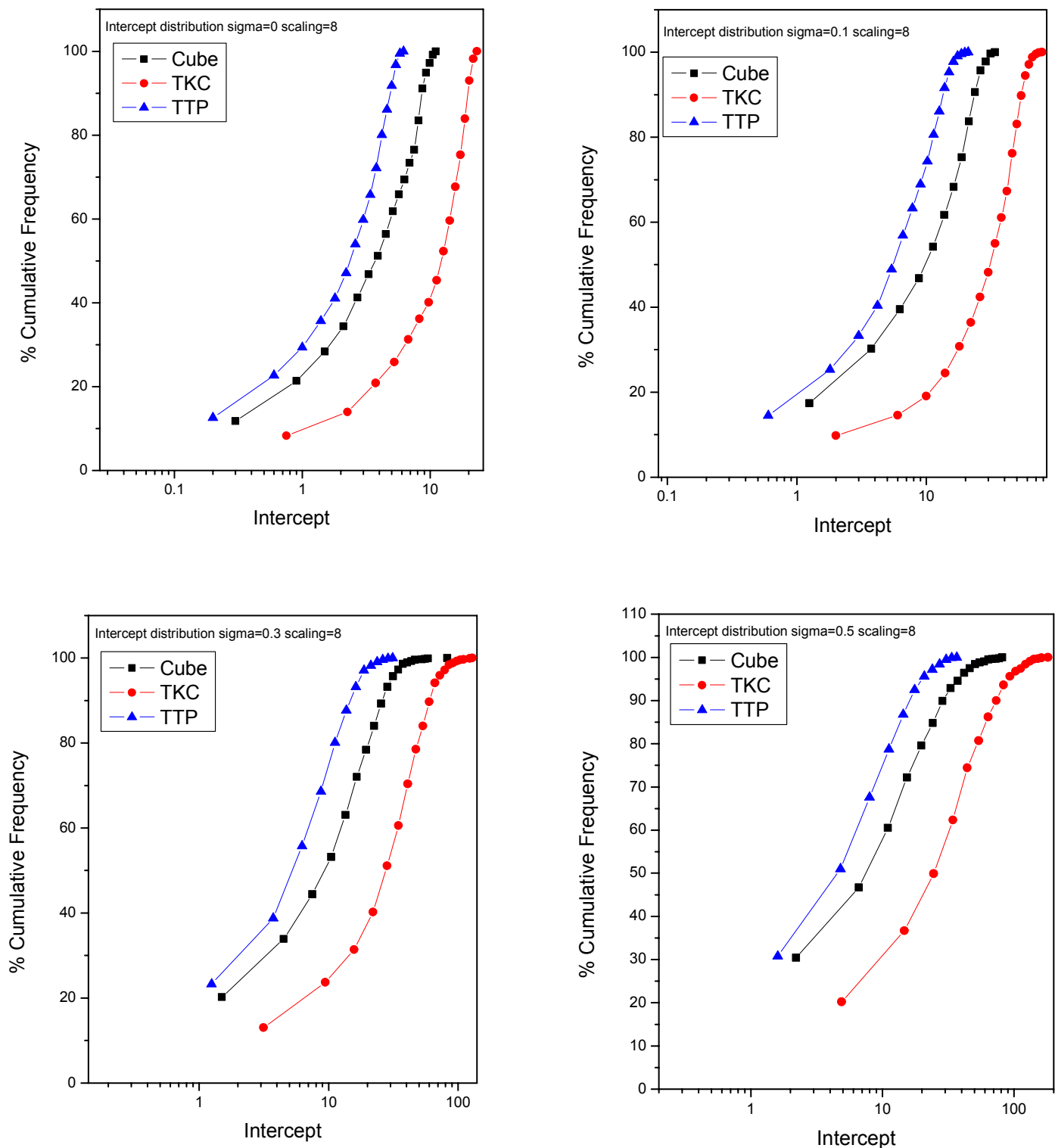


Fig 8 Computed cumulative number distribution plots for intercepts through each crystal shape showing variation with distribution width at a scaling factor of 8.

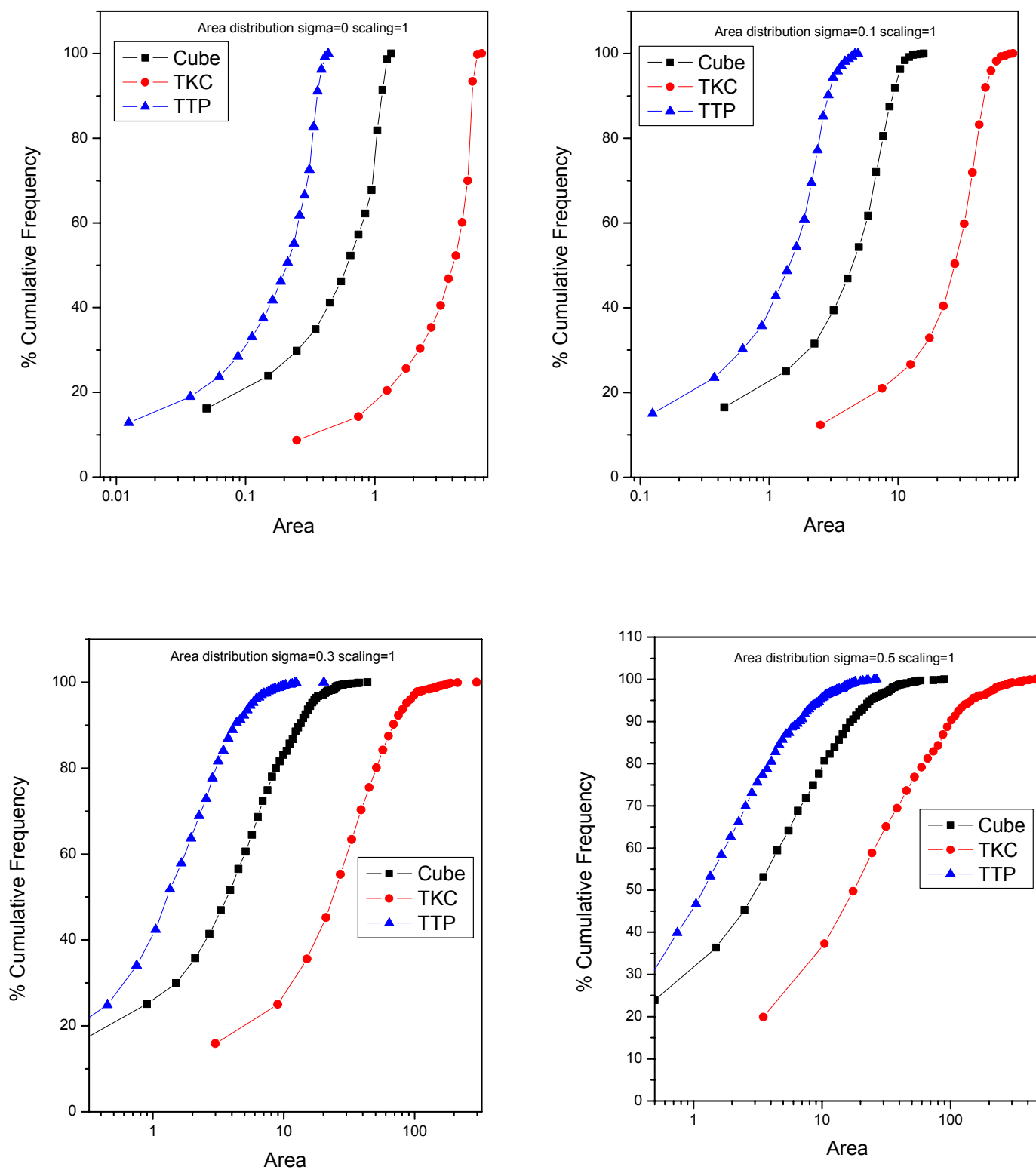


Fig 9 Computed cumulative distribution for areas from each crystal shape showing variation with distribution width for a scaling factor of 1.

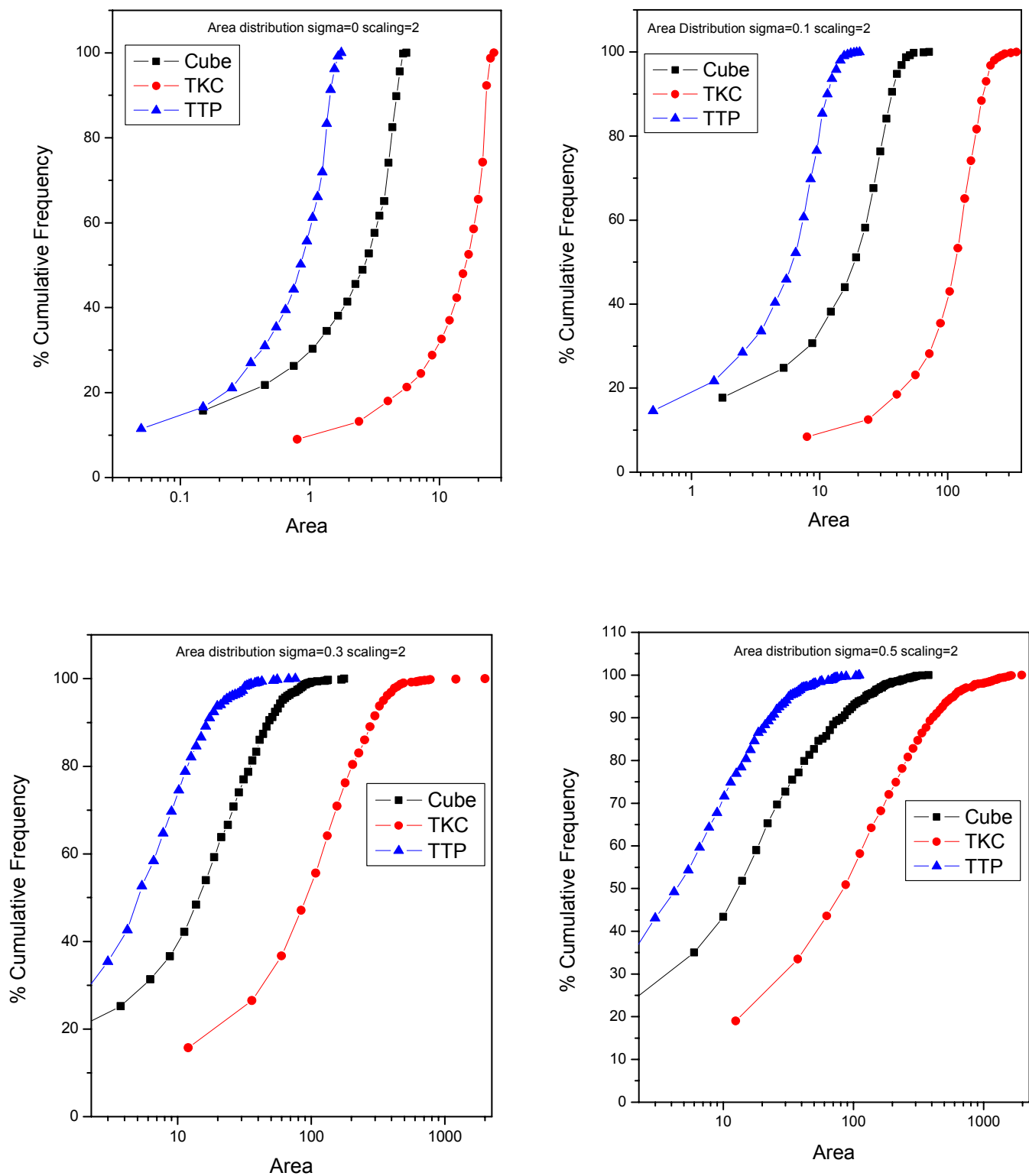


Fig 10 Computed cumulative distribution for areas from each crystal shape showing variation with distribution width for a scaling factor of 2.

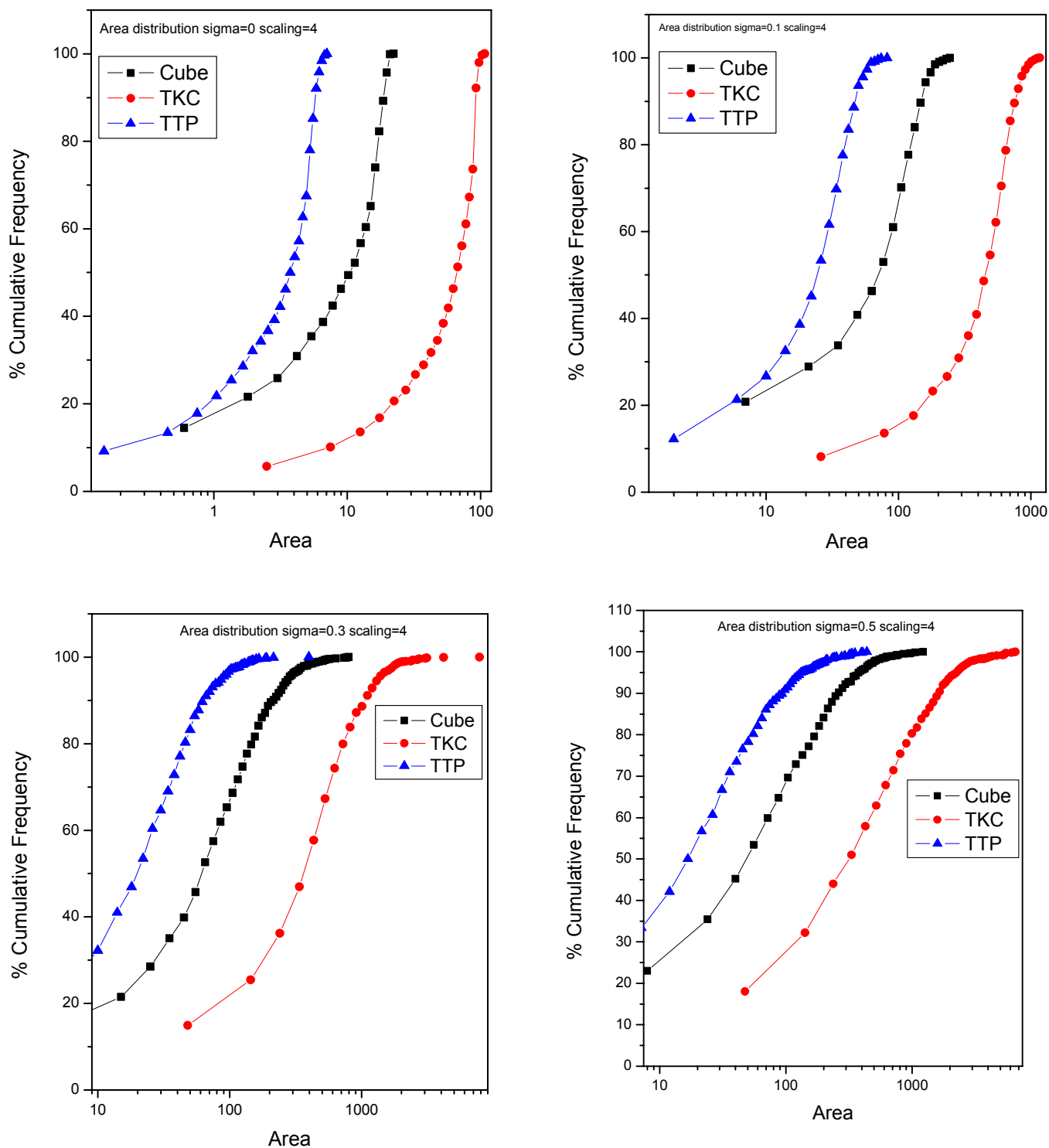


Fig 11 Computed cumulative distribution for areas from each crystal shape showing variation with distribution width for a scaling factor of 4.

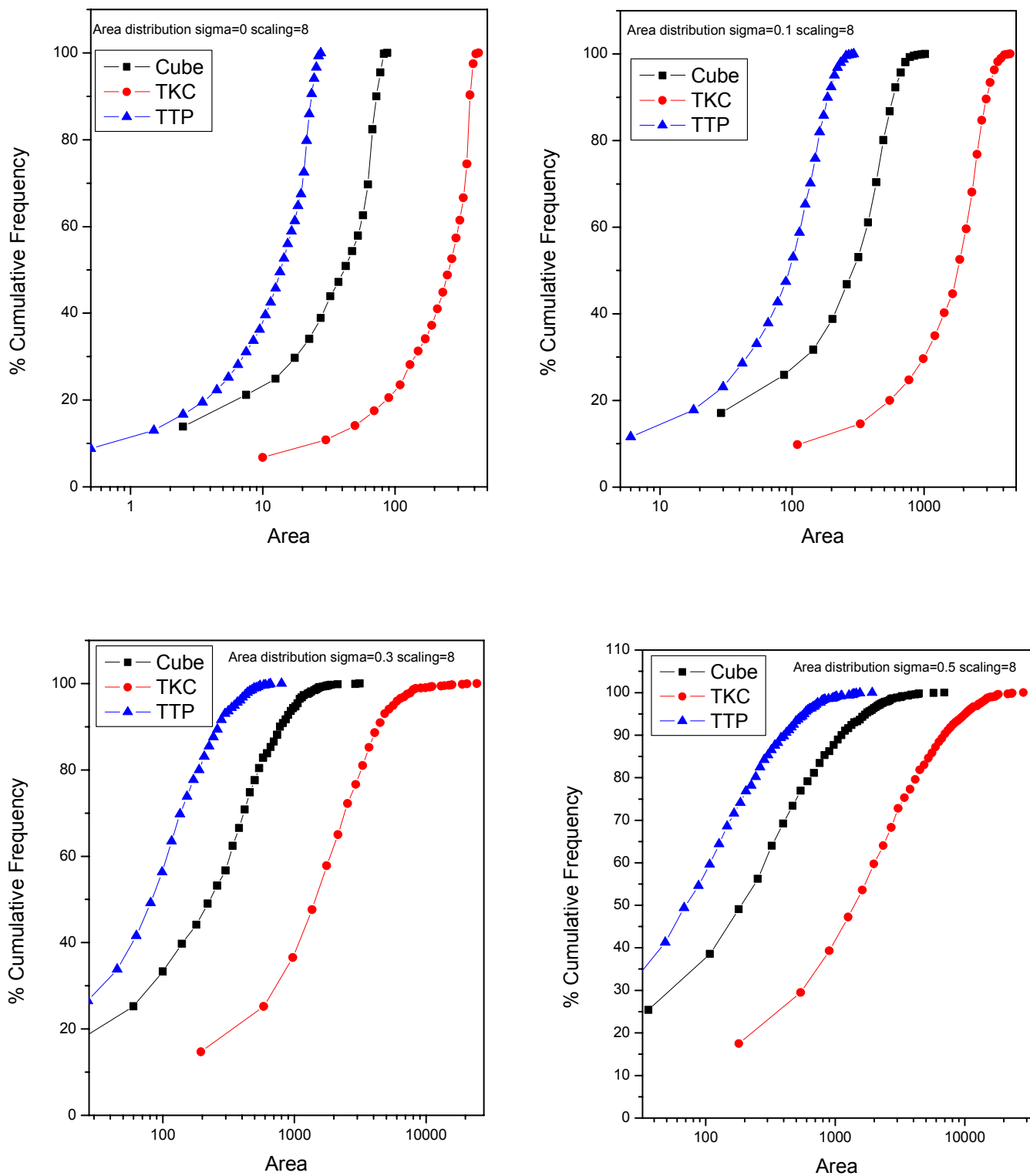


Fig 12 Computed cumulative distribution for areas from each crystal shape showing variation with distribution width for a scaling factor of 8.

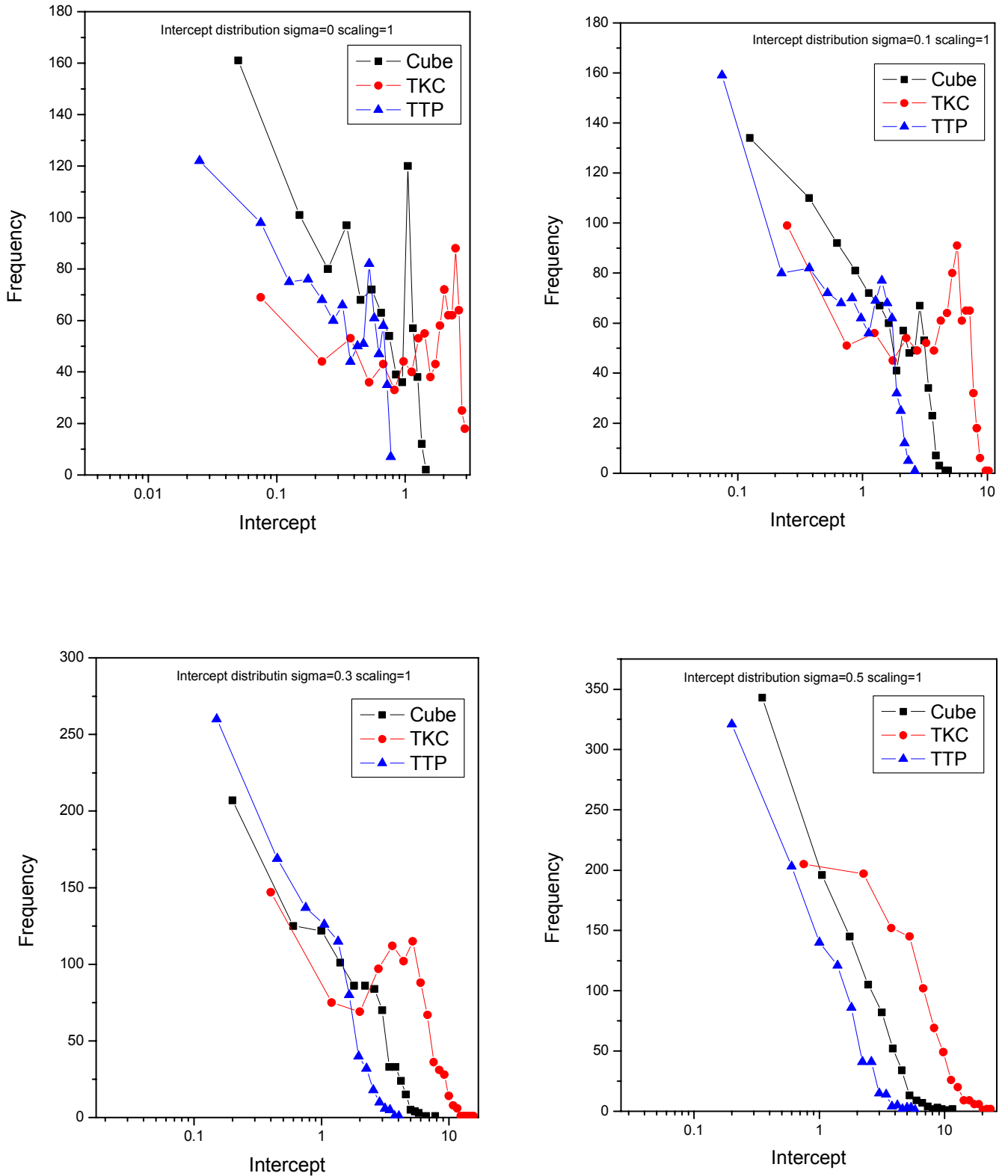


Fig 13 Computed frequency distribution plots of intercepts through each crystal shape showing variation in distribution width at a scaling factor of 1.

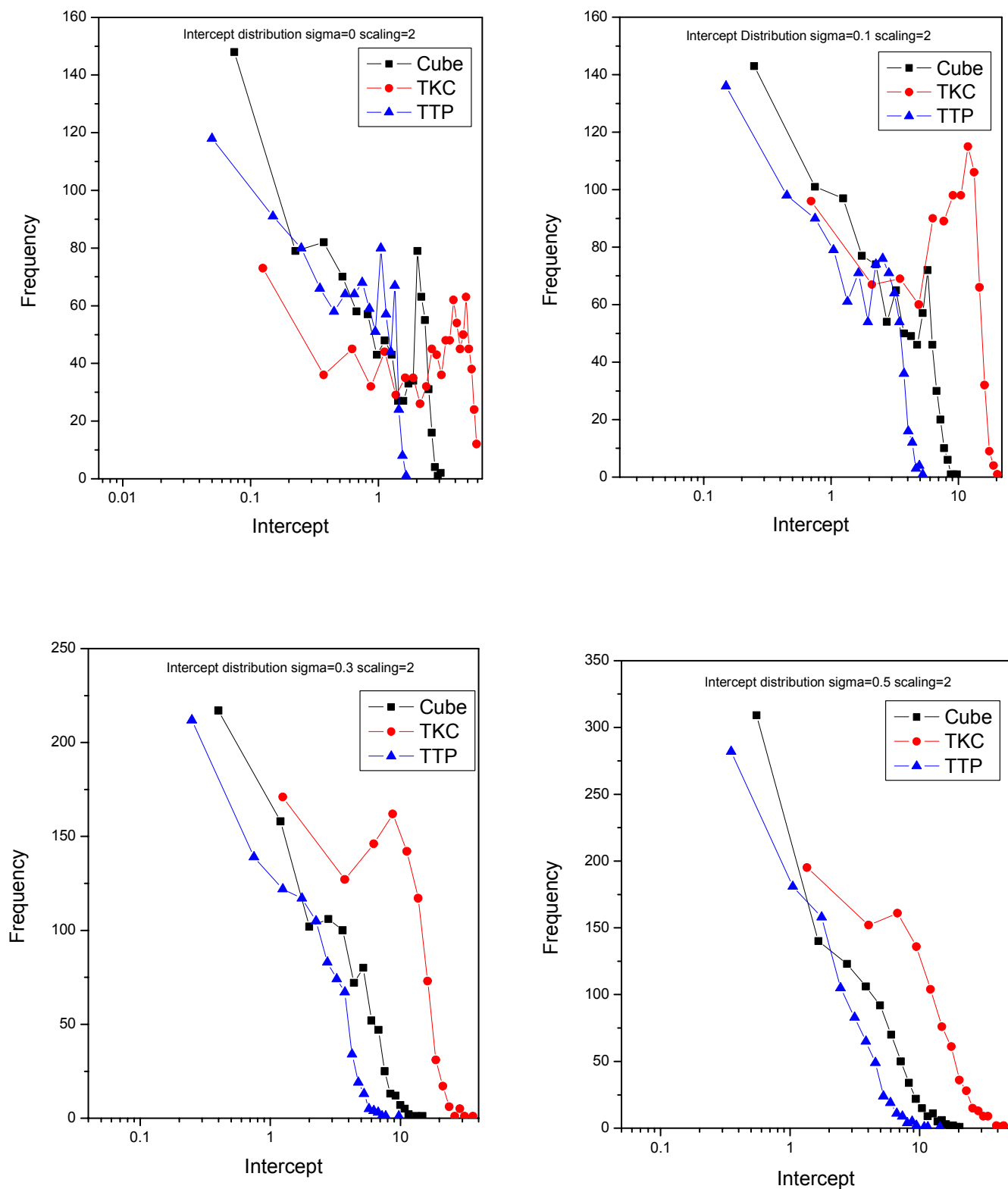


Fig 14 Computed frequency distribution plots of intercepts through each crystal shape showing variation in distribution width at a scaling factor of 2.

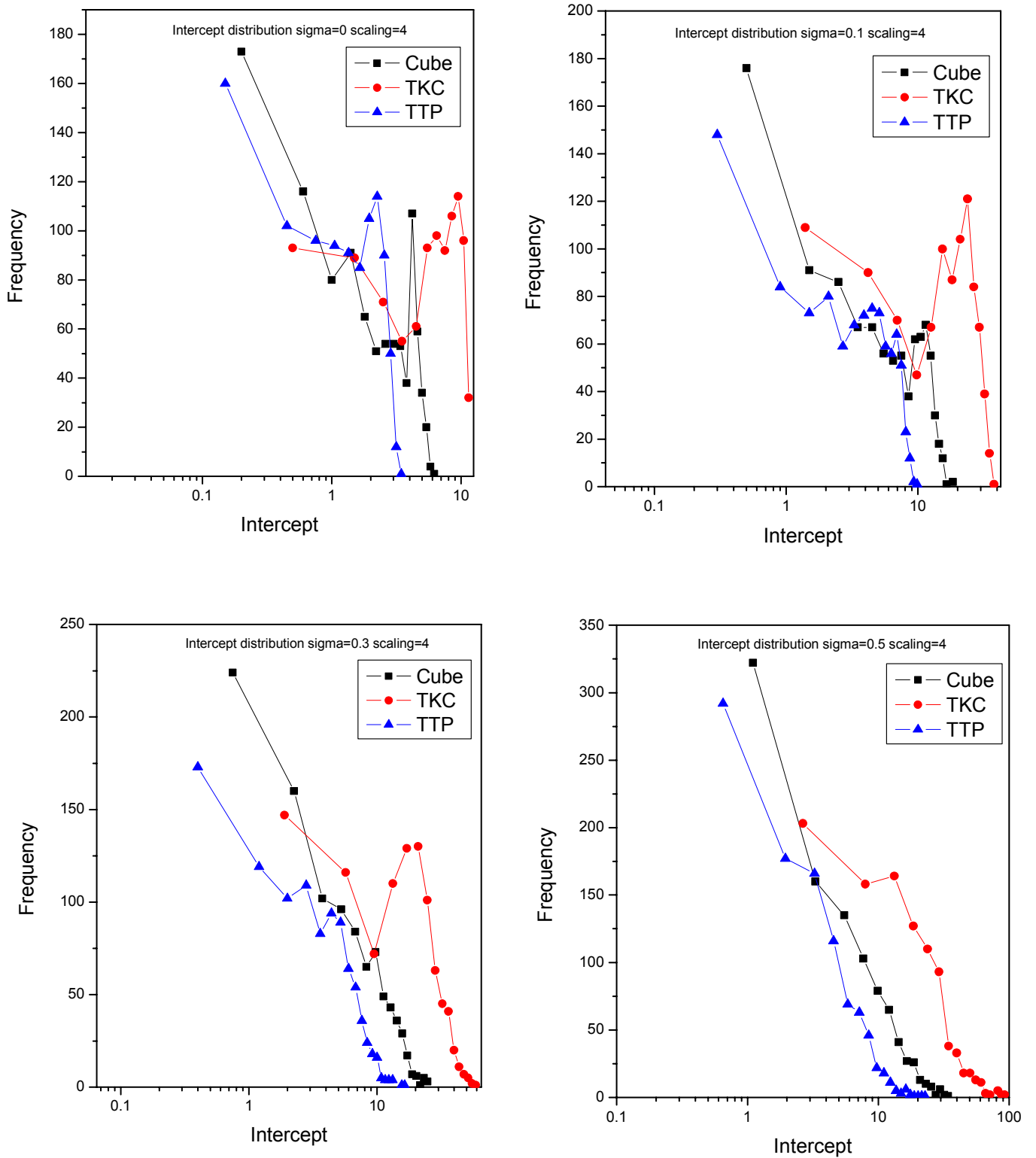


Fig 15 Computed frequency distribution plots of intercepts through each crystal shape showing variation in distribution width at a scaling factor of 4.

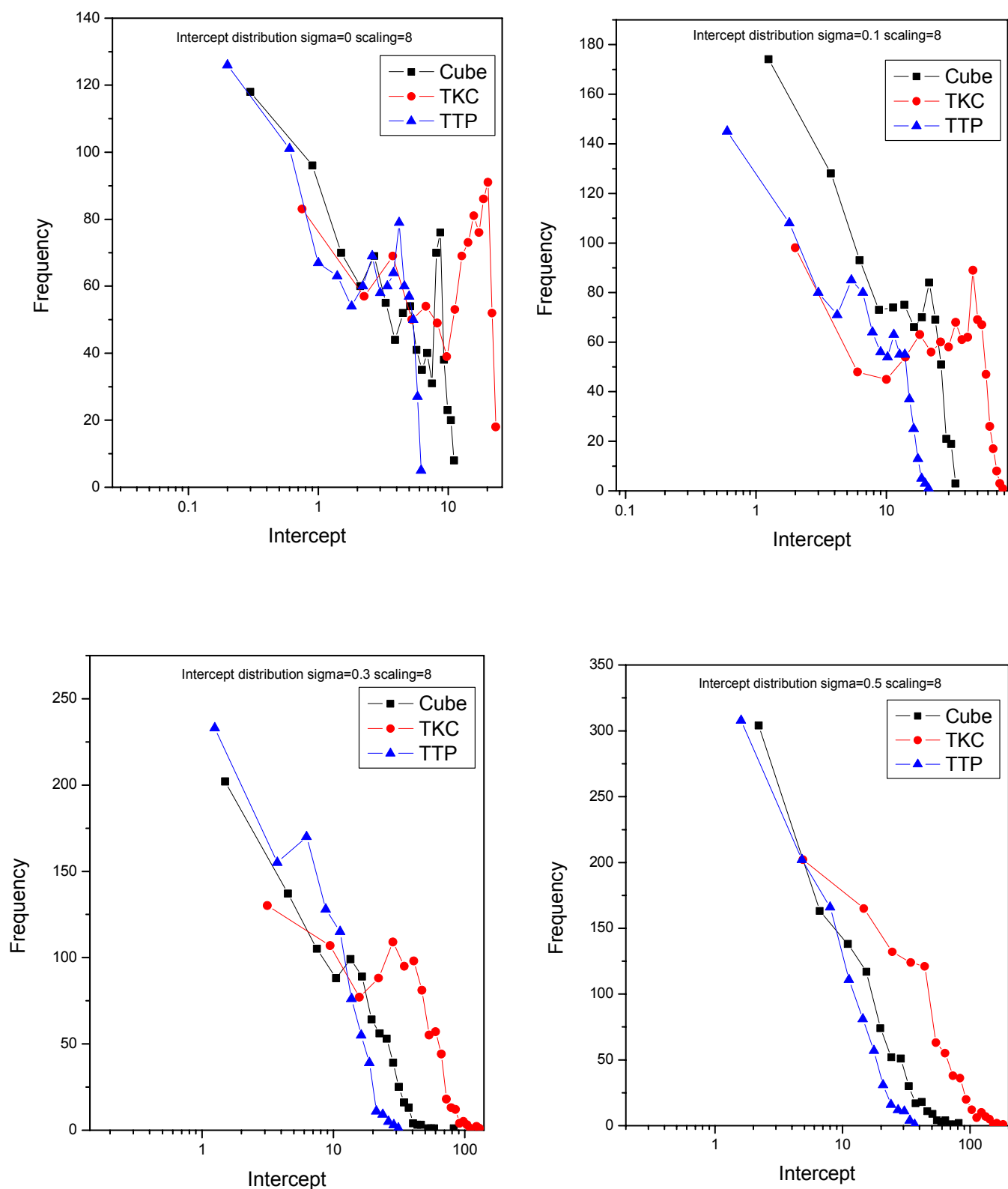


Fig 16 Computed frequency distribution plots of intercepts through each crystal shape showing variation in distribution width at a scaling factor of 8.

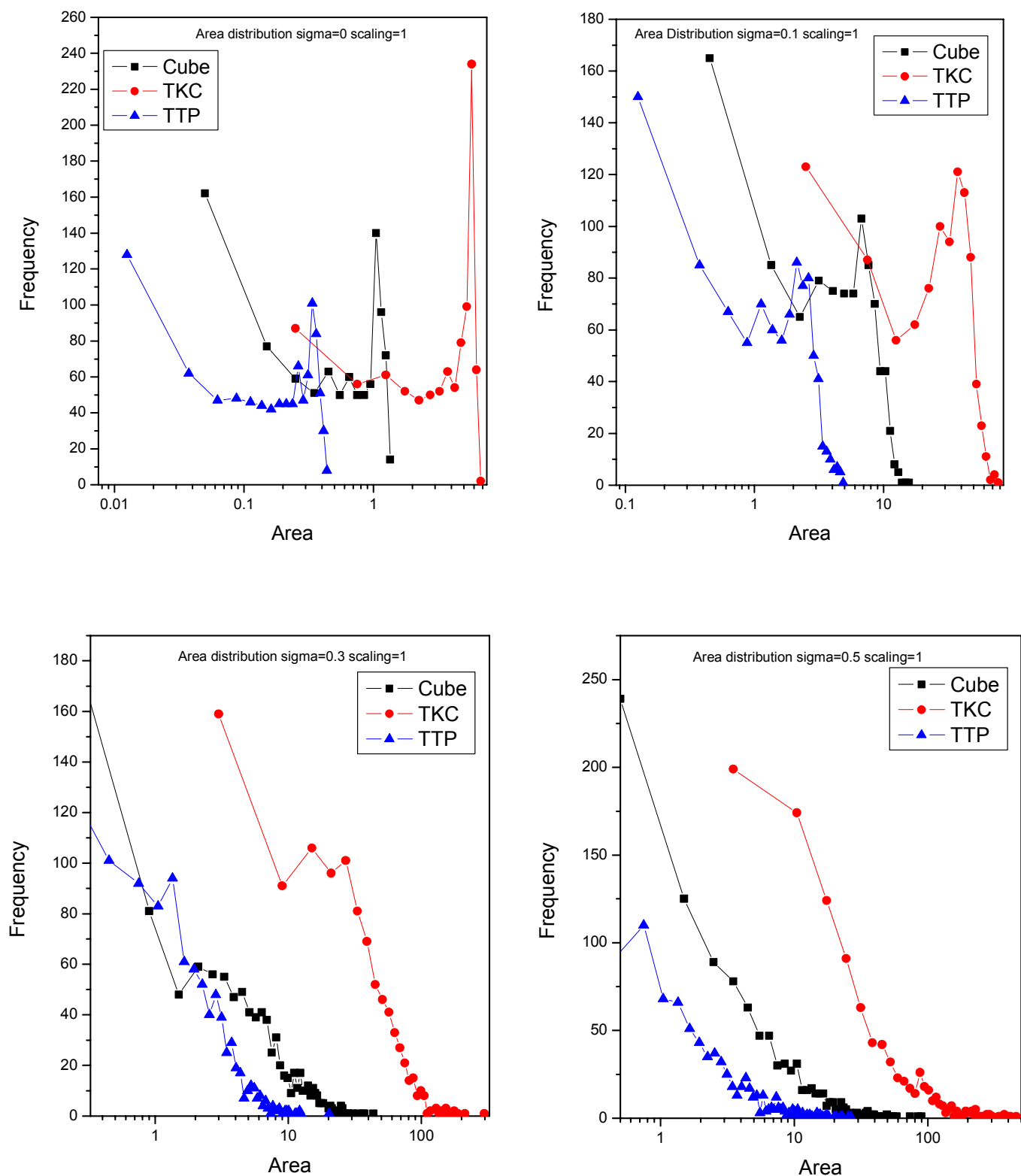


Fig 17 Computed frequency distribution plots of areas through each crystal shape showing variation in distribution width at a scaling factor of 1.

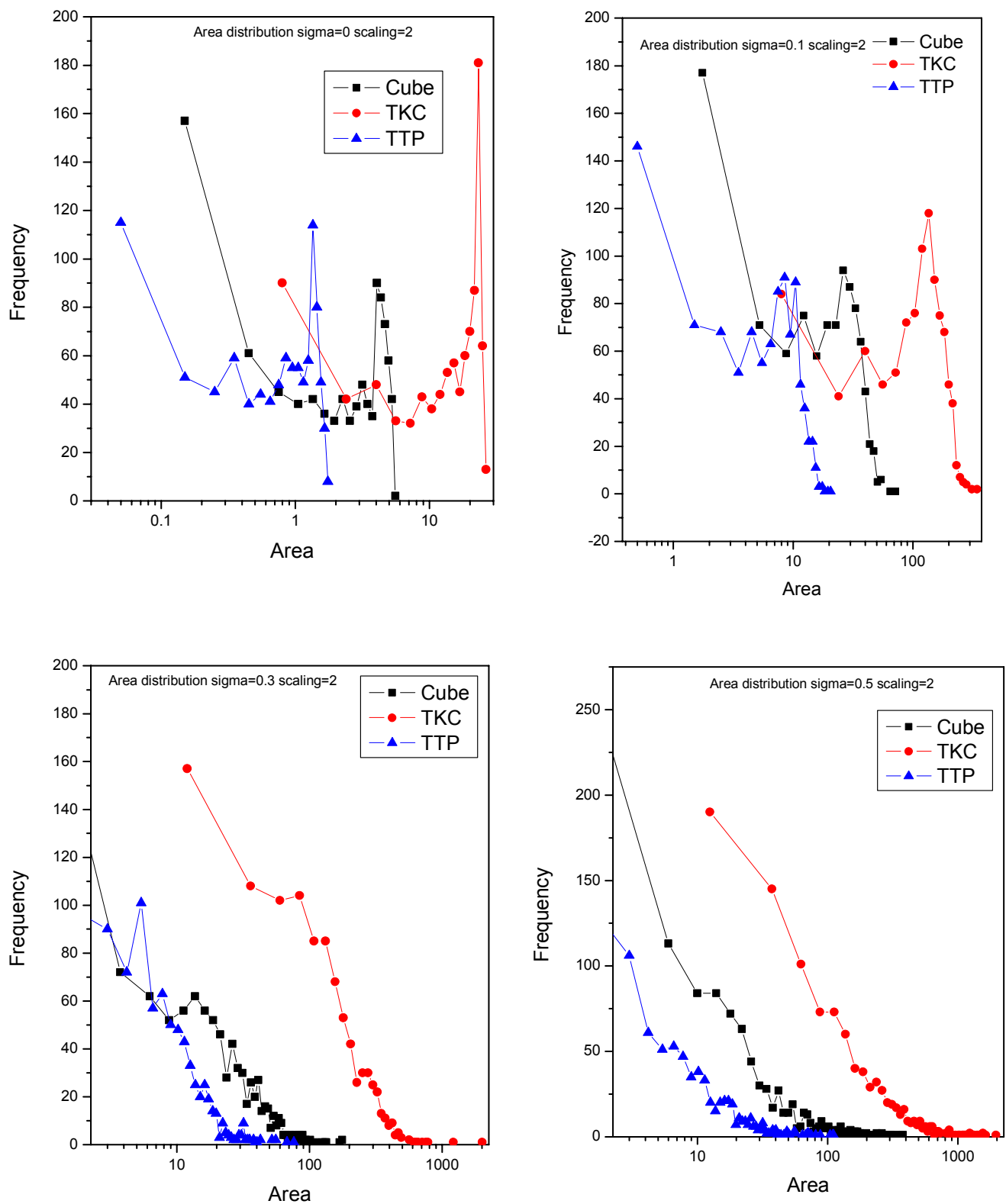


Fig 18 Computed frequency distribution plots of areas through each crystal shape showing variation in distribution width at a scaling factor of 2.

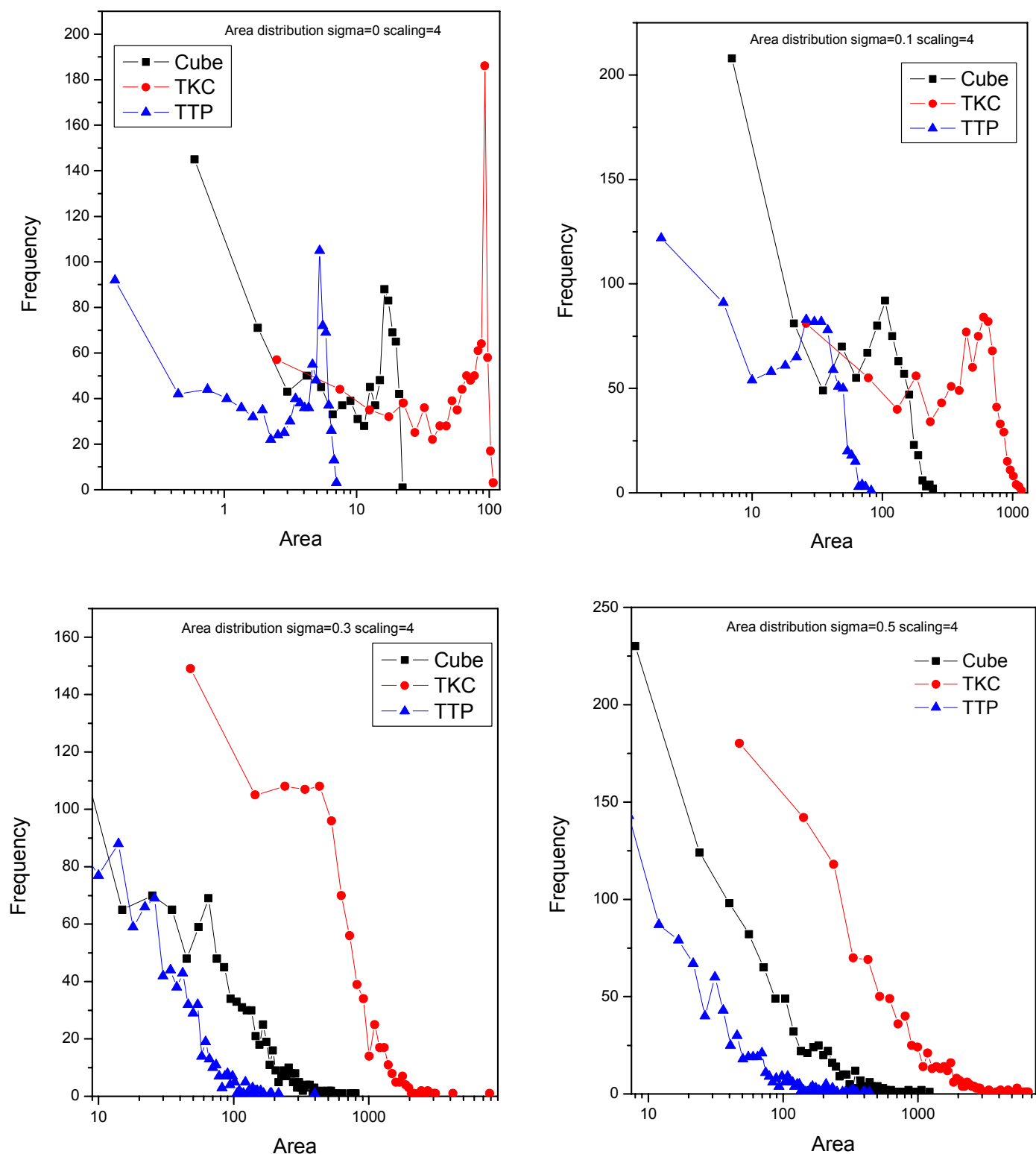


Fig 19 Computed frequency distribution plots of areas through each crystal shape showing variation in distribution width at a scaling factor of 4.

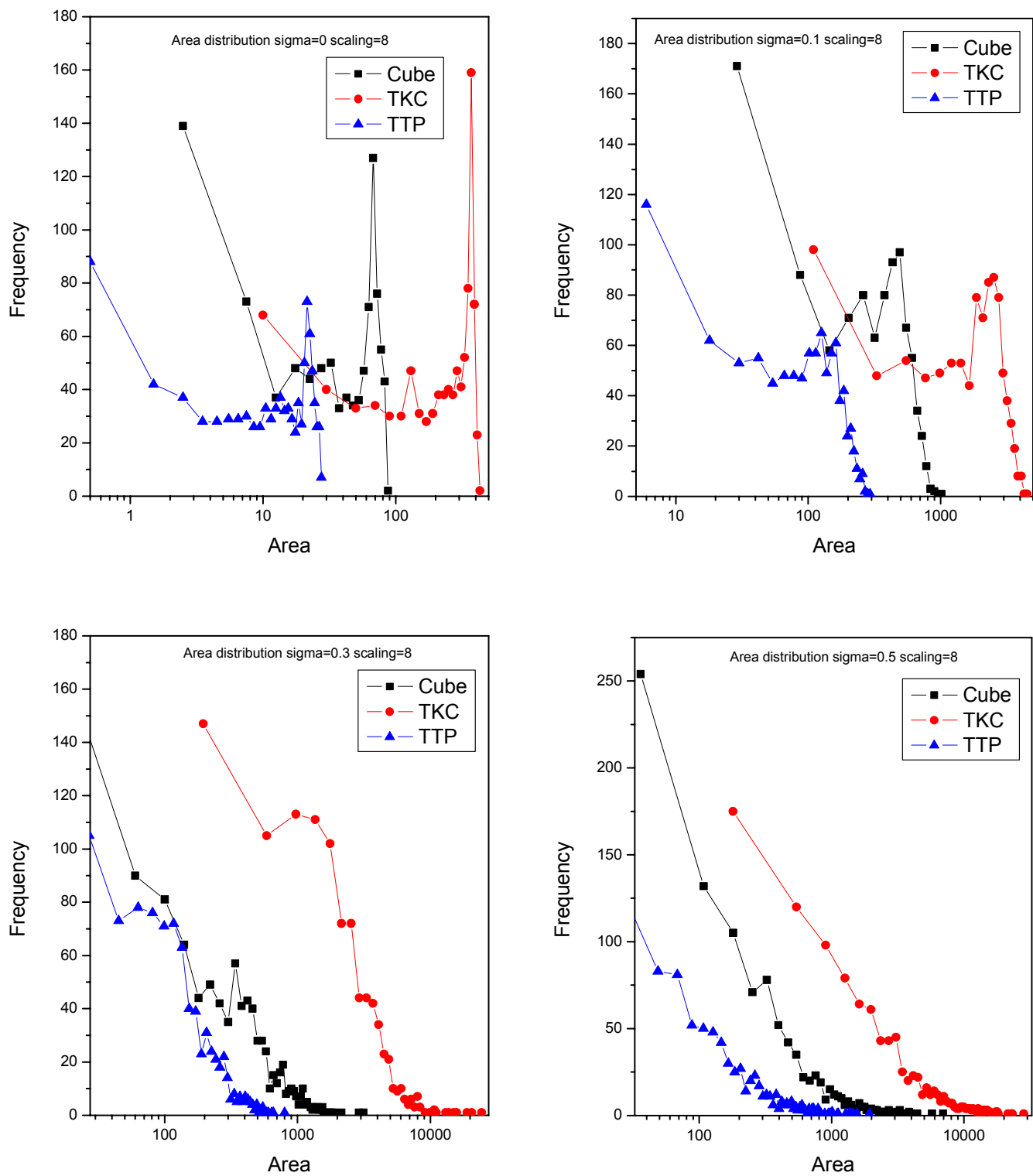


Fig 20 Computed frequency distribution plots of areas through each crystal shape showing variation with distribution width at a scaling factor 8.

Arithmetic mean values for LI and ECD were obtained from the number distribution plots of intercepts and areas (Figs 21-22) and the computed relationship between intercept and ECD appears to be more or less independent of the spread of crystal size (sigma). The pattern that seems to be emerging is that the ECD is about 1.7 times the intercept value. This holds for all values of sigma. Fig 22 also includes a graph showing the affect of computed sample size (between N=100 to 100,000) on the relation between ECD and LI for the TTP shape. There is some evidence that the measured slope is different, slightly higher, for the smallest sample size (N=100), but all the results are quite close.

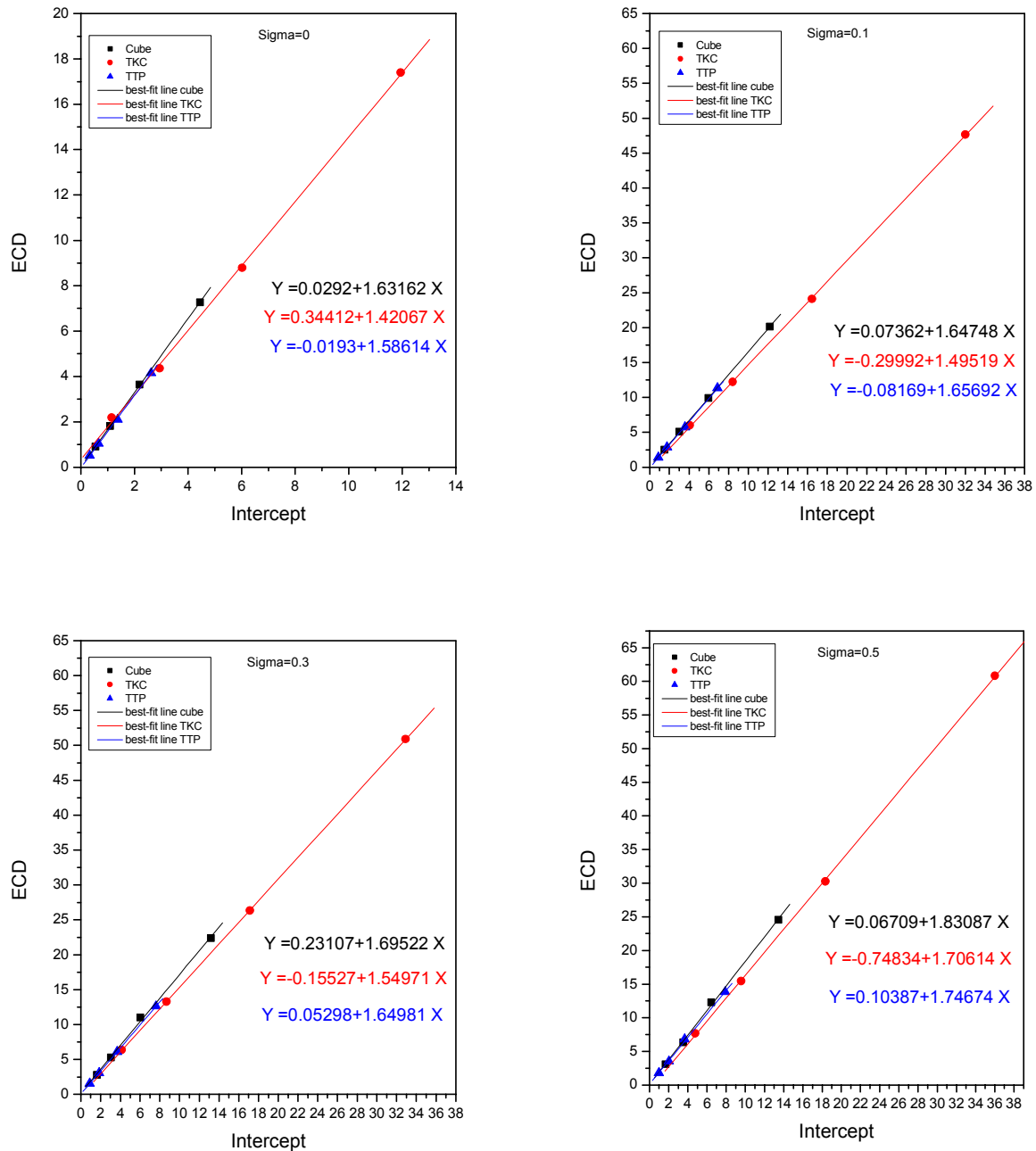


Fig 21 Effect of crystal shape and distribution width on the relation between ECD and LI.

In addition to the relationship between LI and ECD being almost independent of the size distribution of crystals, it appears that it is also almost independent of the shape of the crystal. This is shown by the similarity of the slopes of the plots of ECD against LI for the different shapes with different distributions.

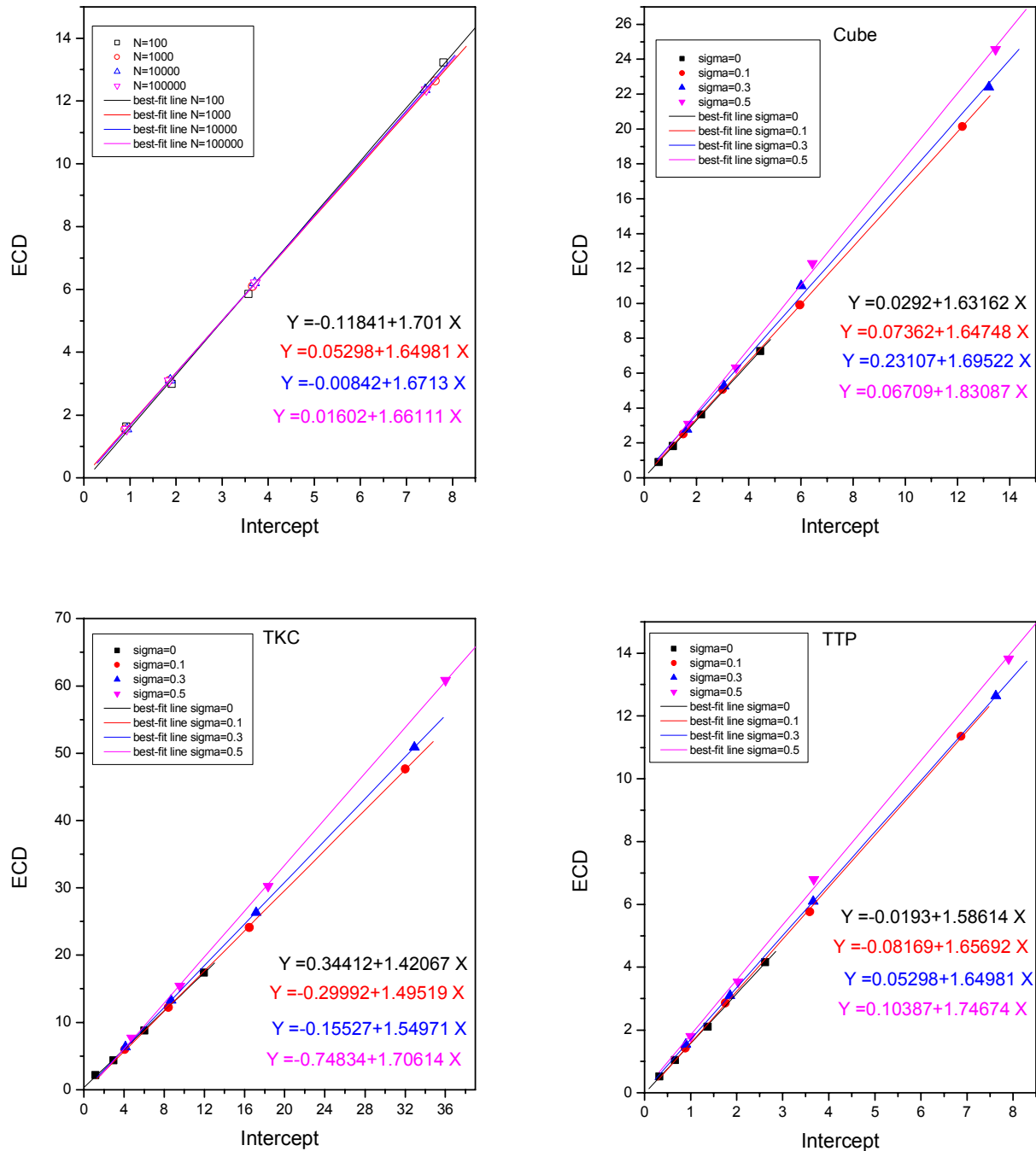


Fig 22 Effects of sample size on shape TTP and distribution width on the relation between ECD and LI for all crystal shapes.

The plot of the slopes of the ECD/LI graphs against sigma for each of the shapes illustrates the similarity between the values. It also shows a trend that as sigma increases the slope of the plot increases (Fig 23). This is true for all the shapes. The anomalous point in the TTP plot is most probably due to statistical variation. Although the plot shows that as sigma increases the slope of the ECD against LI graph increases for all three shapes, it seems to indicate that the relationship between sigma and the slope is slightly different for each shape. The TKC plot could be approximated by a straight line, whereas the TTP and cube plots, ignoring anomalous points, might be better approximated by a curve, possibly exponential in shape.

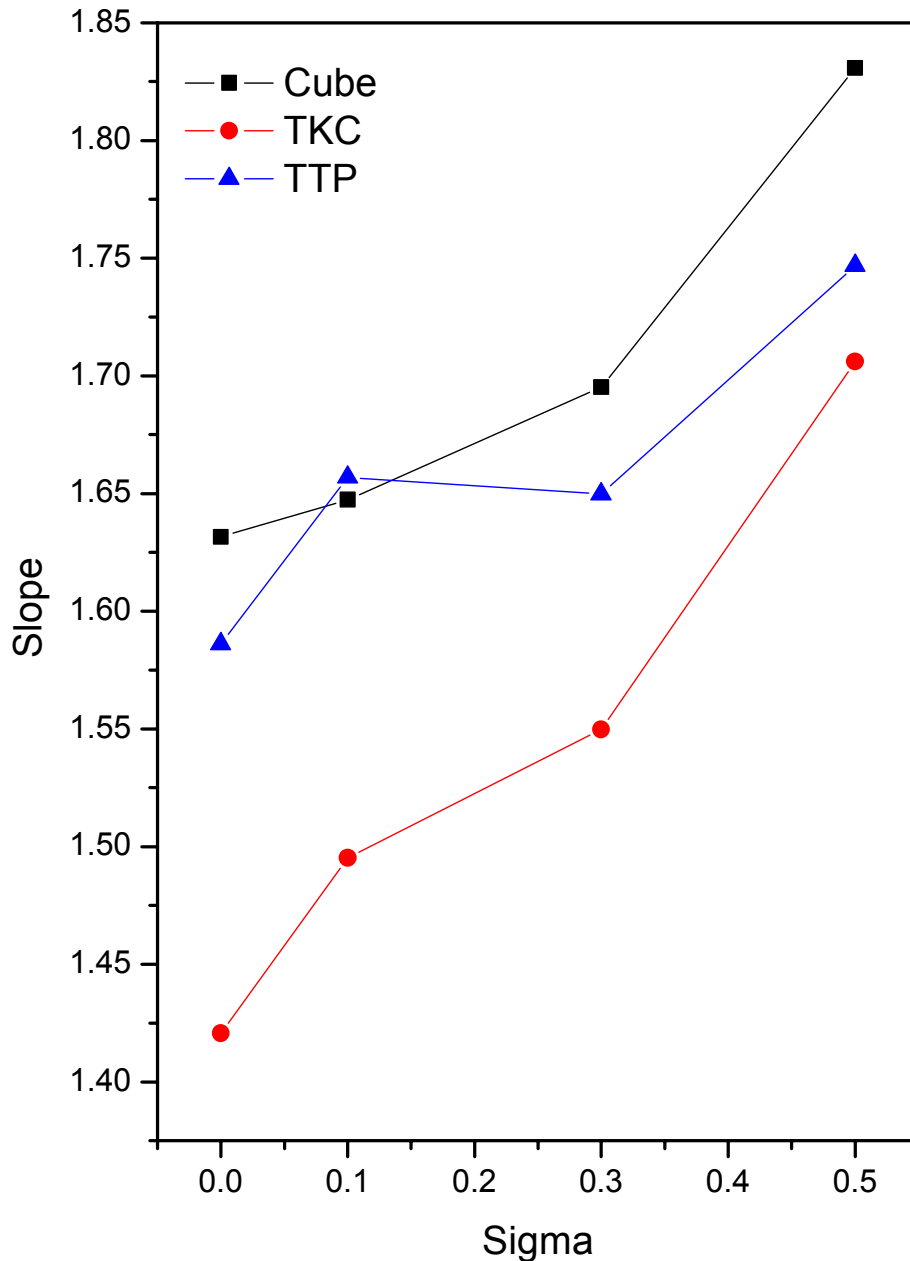


Fig 23 Dependence of shape of ECD/LI plot on distribution width

3.2.2 Effect of TTP Truncation and Shape

A further set of results were obtained by changing the height/length of the TTP crystal for different amounts of truncation (Fig 24). The results are shown in Figs 25-27, including linear best fits.

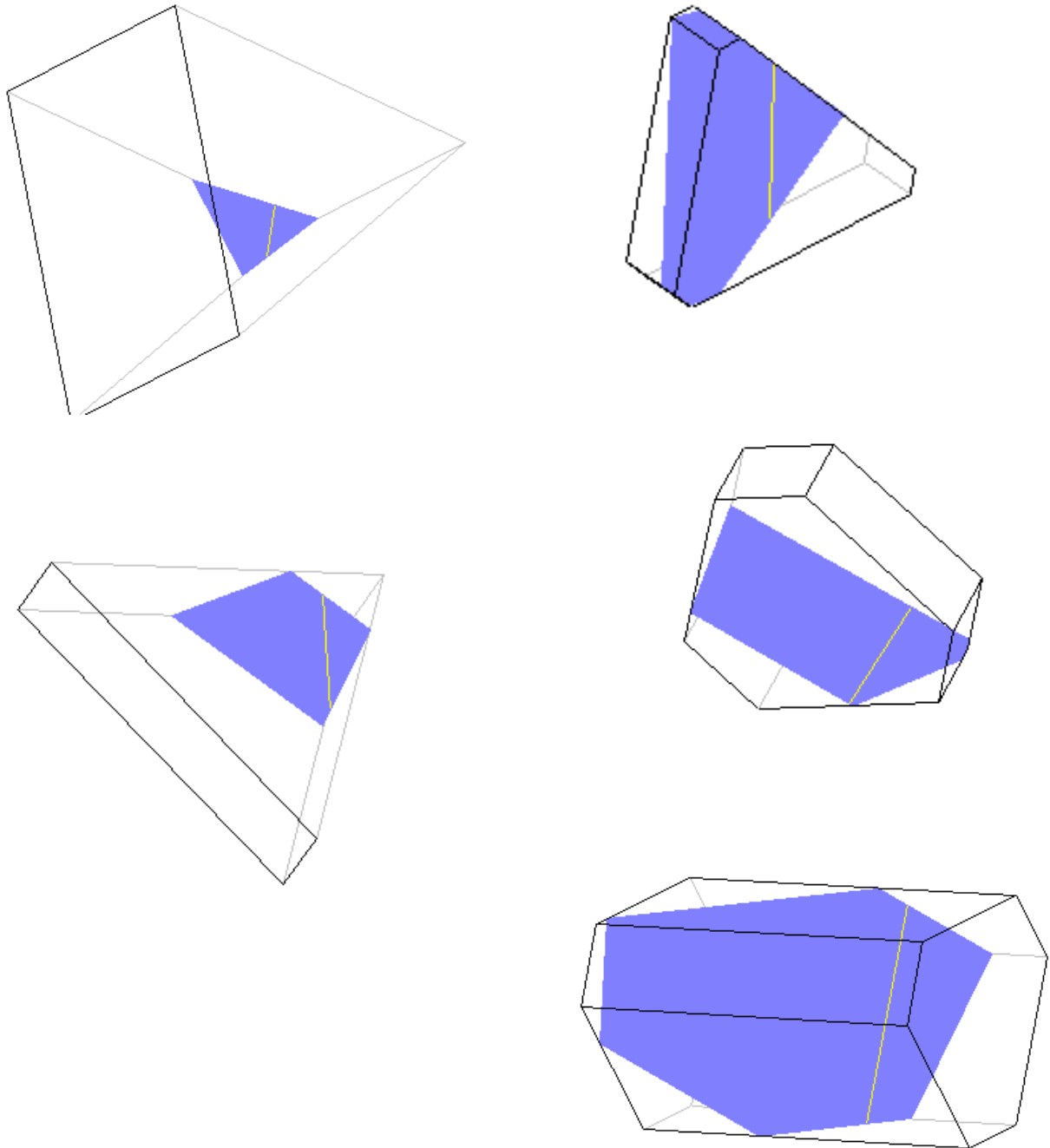


Fig 24 Some examples of different shaped TTPs

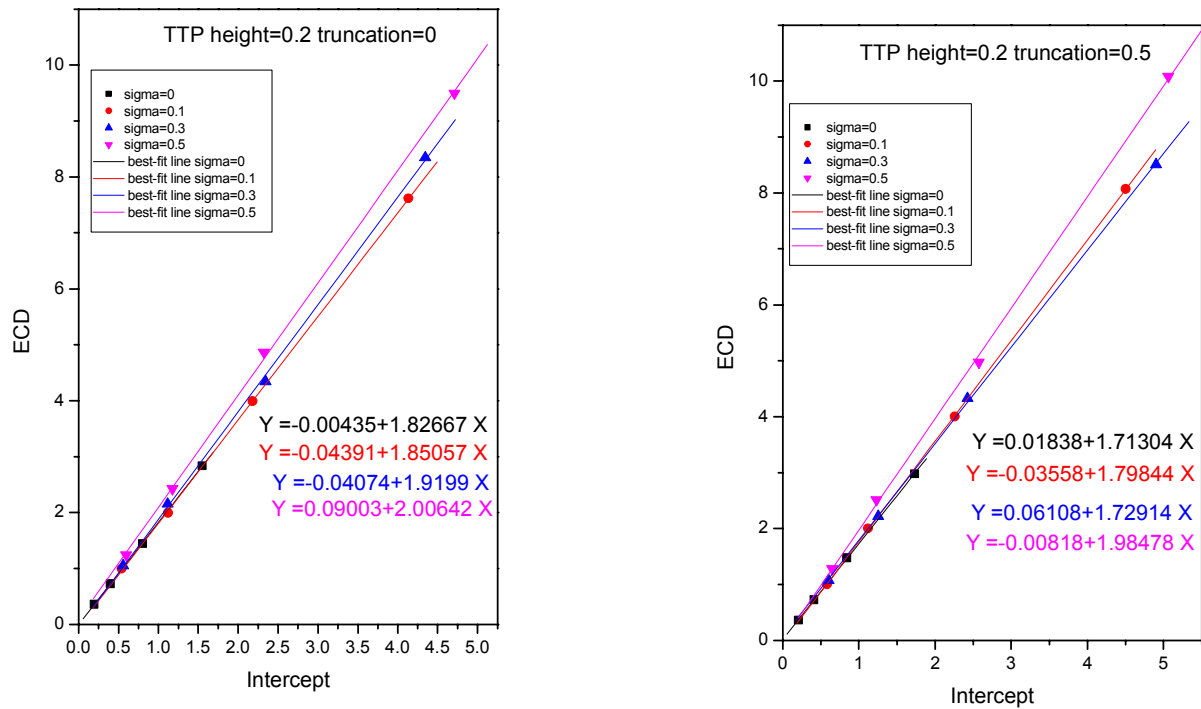


Fig 25 Effect of TTP truncation for a height of 0.2

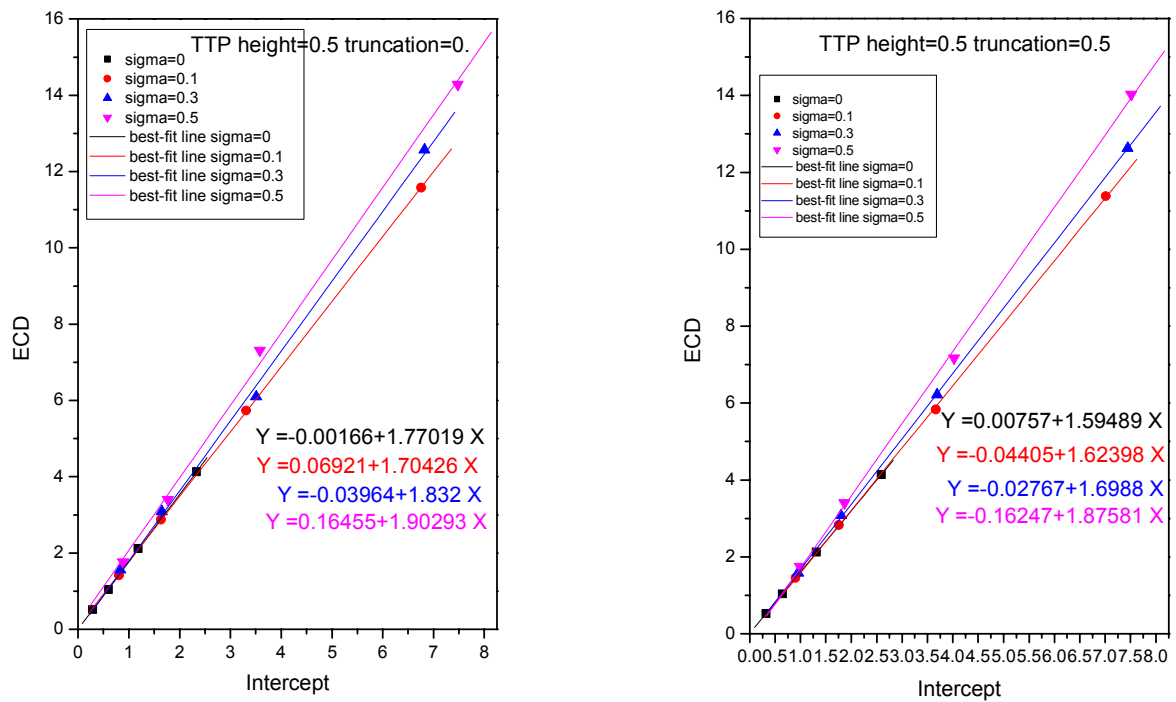


Fig 26 Effect of TTP truncation for a height of 0.5

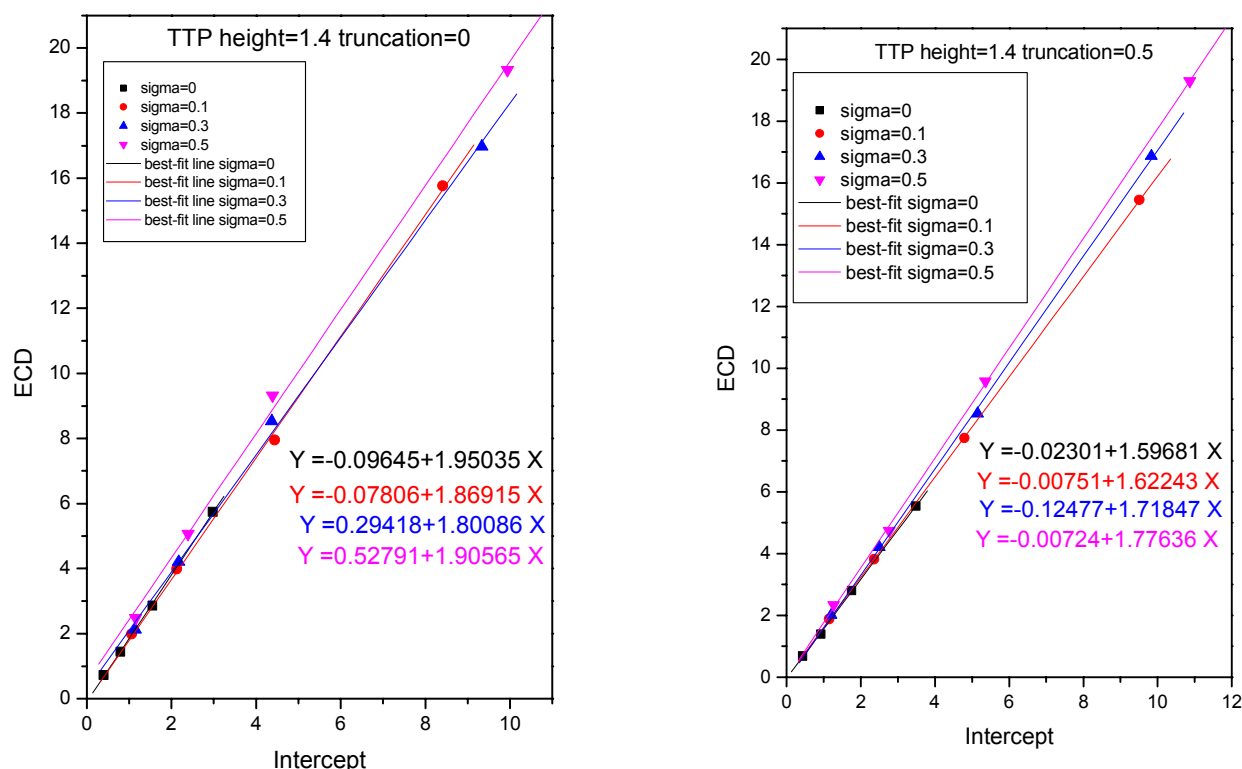


Fig 27 Effect of TTP truncation for a height of 1.4

These simulation studies showed that the shape of the crystal had only a small effect on the relationship between LI and ECD, and also showed that the amount of truncation and the relative dimensions of the TTP shape crystal had little effect. The range of values for the slopes were similar to the range for the different shapes, indicating the similarity between changing the shape and the relative dimensions of the TTP. They both seem to amount to the same change in LI/ECD relationship. This is not unsurprising since changing the height and truncation of the TTP does effectively change its shape, thus you would expect a similar pattern to be found.

The graph of slope of ECD/LI plot against sigma (Fig 28) doesn't show quite the same pattern as that found between the 3 initial shapes, that the slope increases as sigma increases, but it does indicate that as the truncation decreases the slope increases.

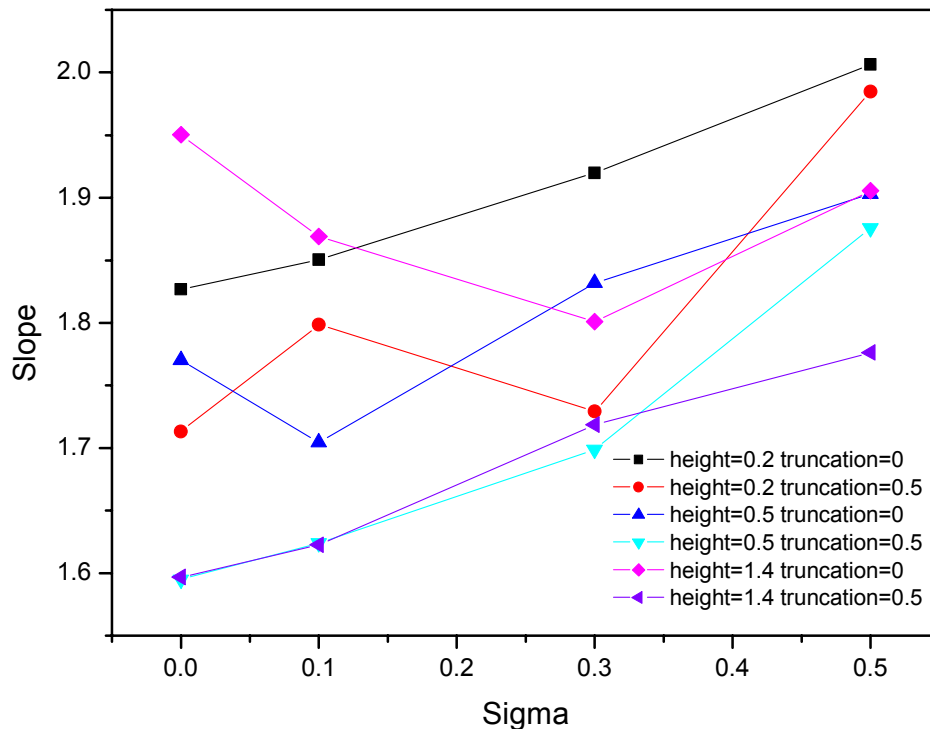


Fig 28 Effect of crystal shape on relation between ECD and LI and distribution width

3.2.3 Effect of Measurement Resolution

When measurements of crystal area and linear intercept are taken from images of sectioned and polished samples they are generally found to follow a lognormal distribution [2,3,11].

However the model produced by POLYCHOP didn't generally follow this trend (Fig 29), where on the left the cumulative frequency is plotted on a probability scale showing deviation from linearity for computed data (taken from Fig 5). On the right hand side experimental data taken from NPL data [8,9] is shown with a good linear fit and no curvature at small values. This difference could be attributed to the accuracy with which practical measurements can be taken compared to that with which the simulation can model. When measurements are taken practically the smallest crystal visible is related to the resolution of the image. This means that when the number of crystals in a given area are counted, the smaller crystals may be under-represented in the distribution as they often cannot be seen. This leads to the average area or intercept being greater than expected, as fewer crystals are counted.

In order to verify the accuracy of the model, the effect of the resolution problem was added to the model, and the resulting distributions plotted. In order to simulate the effects of the resolution the area and intercept data for a TTP were taken from POLYCHOP and grouped into number frequency sets. The smallest bins were then removed, starting with 0, then 2,4,6,8 and 10 bins being removed from the data. -The cumulative frequency plots of the remaining data was then plotted (Fig 30). The pattern exhibited by these plots is that the more bins that are ignored, so simulating a lower resolution of image, the more linear the lognormal

plot becomes. This means that although the data produced by POLYCHOP appears not to give the same pattern as practical data, it is really quite similar. However it exhibits a slightly different distribution due to the lack of practical limitations in measuring small grains or intercept lengths.

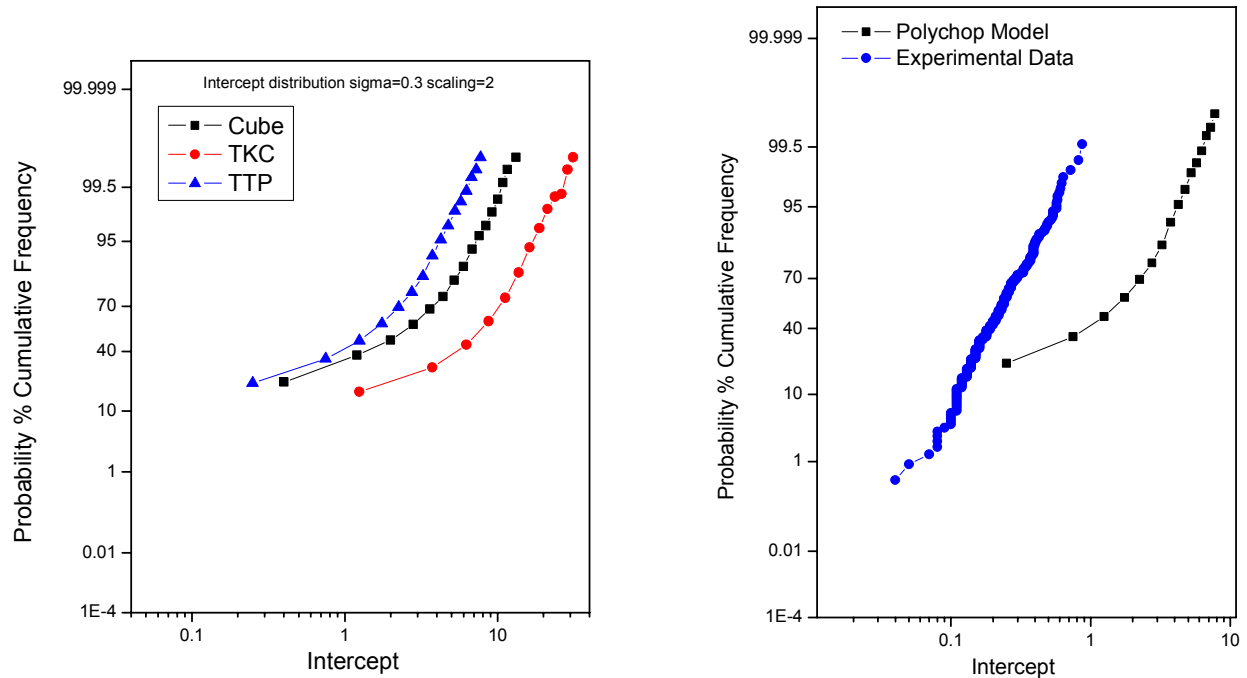


Fig 29 Cumulative frequency plotted on a probability scale.

The knowledge gained from the comparison between the distribution of practical values, and the distribution of modelled values, where no practical limitations apply, can be used as a measure of the accuracy of the practical method. In order for the POLYCHOP data to form a linear pattern 10 bins had to be removed, representing the smallest 10 bin sizes out of 100 bins (10%). This would indicate that the smallest 10% of grain sizes are not measured using the practical method. This value seems higher than expected, and also seems unrealistic in terms of the resolution being probably dependent on the imaging equipment, not on a property of the material being imaged.

The plot of ECD against intercept in Fig 30 for different numbers of bins removed shows that although removing bins affects the distribution of the data it has only a small effect on the relationship between ECD and intercept, which remains linear, and of almost constant gradient.

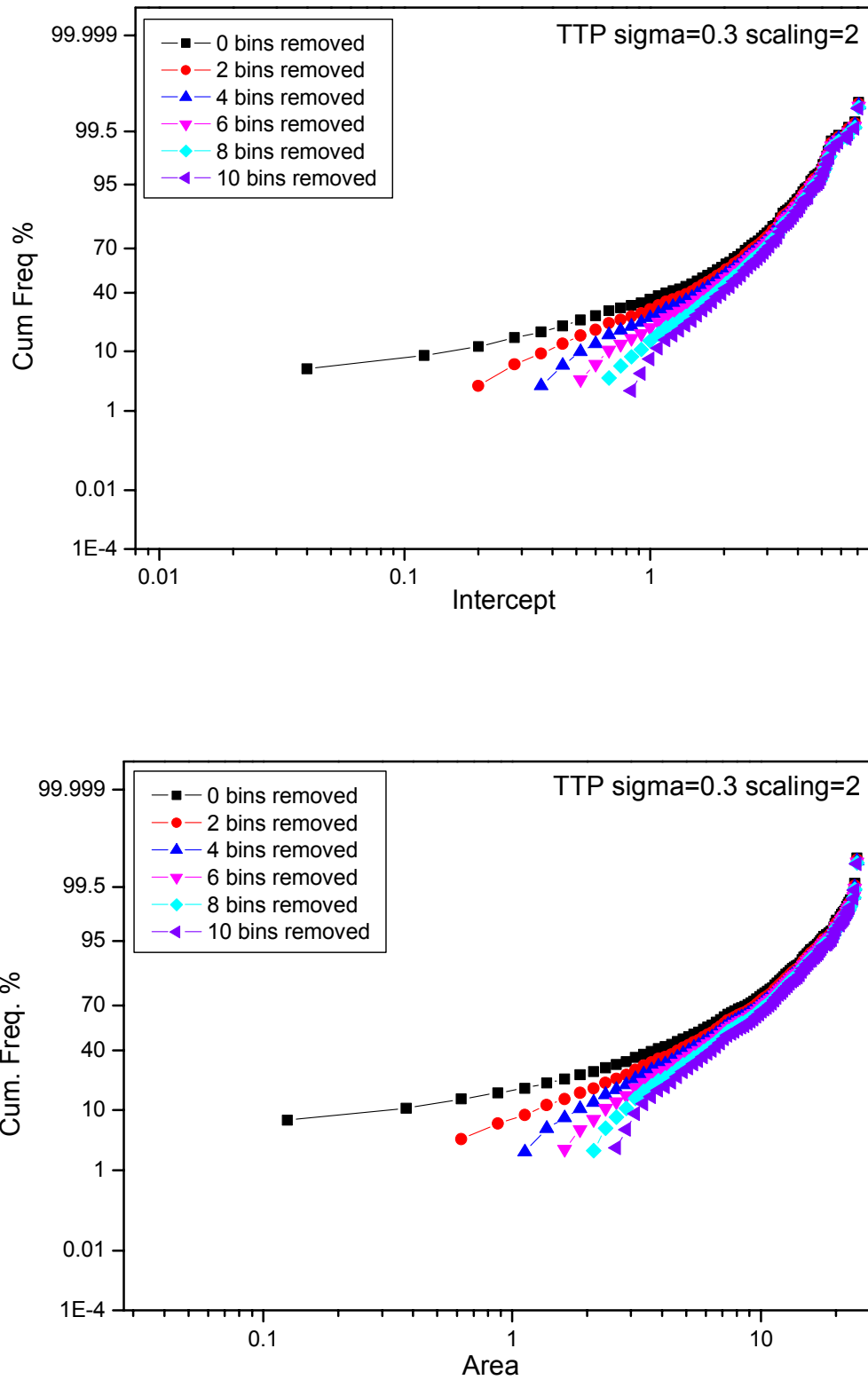


Fig 30 Effect of systematically removing smallest bins from computed distributions of intercept and area

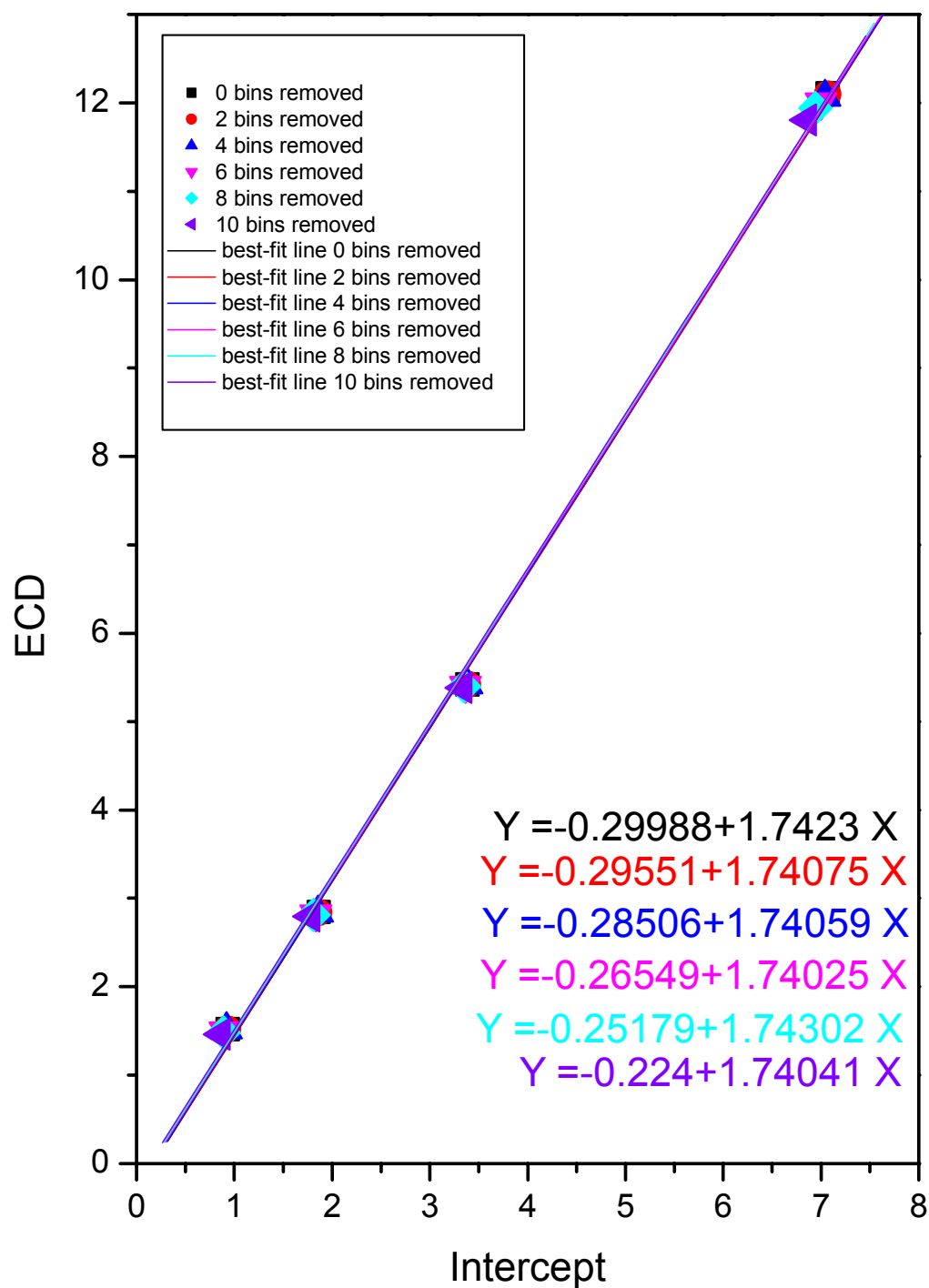


Fig 31 Effect of very small intercepts on the relation between ECD and LI

4 SUMMARY

A study of the relation between the equivalent circle diameter (ECD) and linear intercept (LI) methods for measuring the grain size of WC/Co materials has been conducted through an extensive modelling exercise supported by a limited number of experimental data.

The experimental data indicated that

$$\text{ECD} \approx 1.15 \text{ LI} \quad (1)$$

This is in good agreement with the empirical relation $\text{ECD} = \sqrt{4/\pi} \text{ LI}$ found in measurements on aluminium and ferrous alloys [10]. It would be reasonable therefore to use this expression for converting measurements of linear intercept grain size to equivalent circle diameter grain size and vice versa.

However, the computed data indicated that

$$\text{ECD} \approx 1.7 \text{ LI} \quad (2)$$

At present no satisfactory reason for this difference is known. Further, more detailed experiments will be conducted to check expression (1) above. The modelling studies showed that the relationship between ECD and LI was only slightly sensitive to:

- Crystal shape, for the three shapes studied (cube, truncated triangular prism and tetrakaidecahedron).
- Amount of truncation and aspect ratio of truncated triangular prisms.

Modelling studies indicated that a lack of measurement resolution may affect the detailed characterisation of distribution plots of grain size at smaller values.

5 ACKNOWLEDGEMENTS

The work reported in this document was performed as part of the DTI research programme on the characterisation of powder route materials (MPP6100 – Durability of Hard Materials) in support of the ISO standardisation process for WC/Co hardmetals. Members of the British Hardmetal Research Association are thanked for the supply of materials and discussion of preliminary results. Former NPL colleague Austin Day is thanked for developing the POLYCHOP programme.

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