ACTA STEREOL 1982; 5 - 21
PROC 3RD EUR SYMP STEREOL 2ND PART

THE KEY POSITION OF STEREOLOGY IN QUANTITATIVE MICROSTRUCTURE-CORRELATIONS OF MULTIPHASE MATERIALS

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ABSTRACT

Stereology as part of quantitative microstructural analysis provides the tool to derive "spatial" microstructural parameters from quantities measured in two-dimensional sections. Those parameters form well defined microstructural factors not only quantitatively describing the microstructure of multiphase materials but also being contained in microstructure - field property - equations, which

- open a better scientific insight in the materials behaviour and its dependence on the microstructure
- enable to substitute direct property measurements by calculating the properties from measured microstructural data
- provide a platform to produce tailor-made materials with predicted properties and pre-calculated microstructures to substitute elements, components, alloys and compounds less available in the future.

INTRODUCTION

In materials science the interrelationship between microstructure and properties become more and more important not only for getting a better scientific insight into the material and its behaviour but also for the need to develop tailor-made materials in front of - and to substitute - less available elements in the near future. And a secondary reason for

looking quantitatively at microstructural effects on properties is to find out, whether one may substitute direct property measurements under difficult conditions by a more simple microstructural analysis. This - especially - was the starting point in nuclear materials when considering the porous fuel of a nuclear reactor: on the one hand because it has pores from the sintering process by which it is manufactured, on the other hand pores are created by fission gas formation during reactor operation. These pores migrate following the temperature gradient which exists across the fuel rod radius (fig. 1), altering the thermal conductivity of the material. Due to the irradiation influence - and additionally the high temperature - the variation in thermal conductivity cannot be measured experimentally. The only available sources of information are

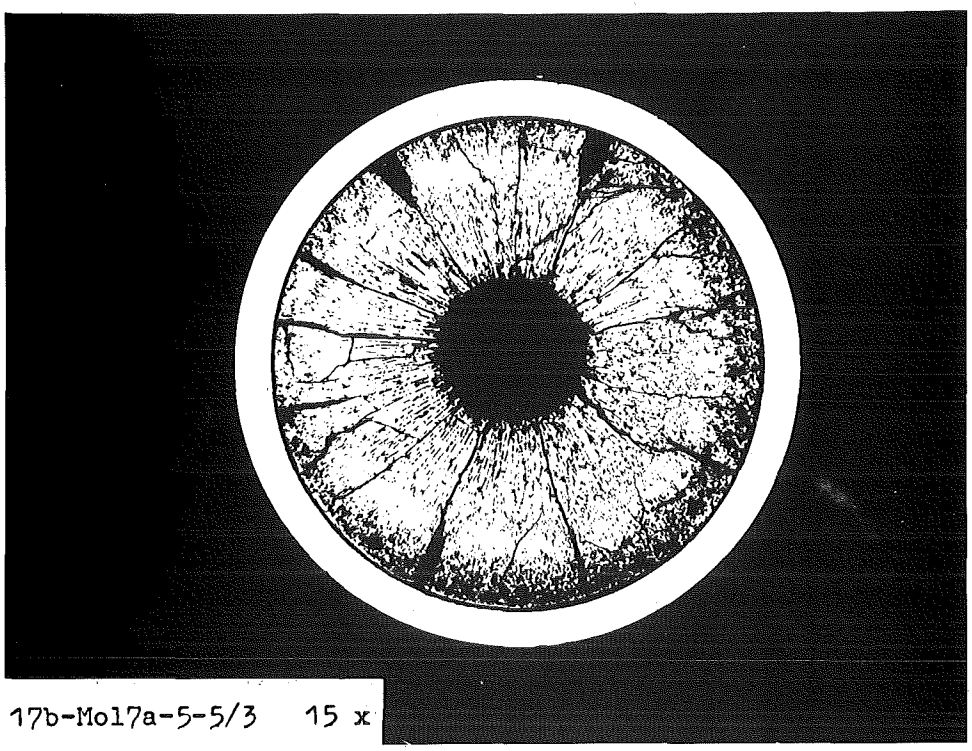

Fig. 1: Cross section of a UO_2 fuel rod cladded by steel

microstructures of the fuel, as shown in fig. 1, obtained after different operation conditions as burnups, by cutting and preparing the fuel rods in hot cells. The central question with respect to nuclear safety is whether the heat transfer from the fuel to the coolant could be decreased by the porosity after a certain time of the reactor life to such an extent that the fuel center enters its melting point. If this would happen, it would be the start of a reactor core melt accident - and it was the start for the work presented here to investigate the effect of second phase inclusions on the properties as the thermal conductivity of a two phase material. In this context pores are only a special case of second phase inclusions.

MICROSTRUCTURE-PROPERTY-EQUATIONS

Compiling the literature one may make use of the analogy between the field equations of all field properties such as thermal and electrical conductivity, dielelectrical constant, and magnetic permeability. There are about 40 equations describing the effect of the microstructure on the field properties of two-phase materials, one of the first derived in 1821. To select those which could form a reliable basis to enter the present problem all equations have been tested under direct plaussible limiting conditions as these for example:

- if the concentration of one phase becomes zero the overall conductivity has to become identical with the conductivity of the other phase
- if the conductivity of one phase approaches infinite the overall conductivity has to remain finite , because this case refers to porous materials

Three equations stood the treatment:

- the Maxwell equation /5/
- the Bruggeman equation /1,2/ and
- the Niesel equation /6/

presenting the following course for deriving the equation of the effective conductivity of a two-phase material as a

function of its microstructure: a homogeneous electrostatic field in a single phased material is assumed as shown in fig. 2.

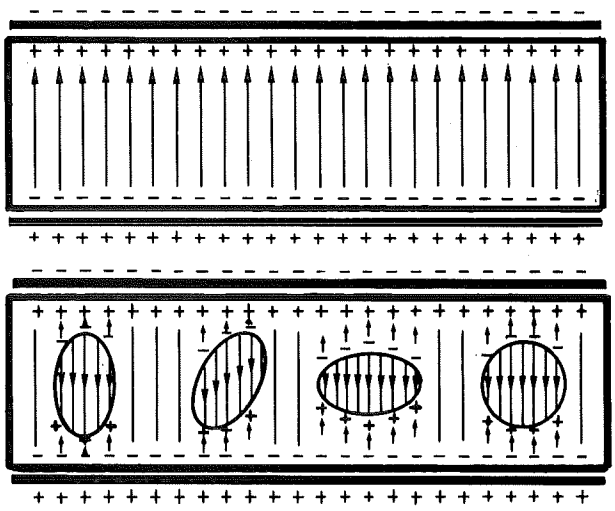

Fig.2: Primary field and stray field superimposition

To consider the effect of a second phase on the properties of a material, one has to introduce second phase particles in it. Here - first time - stereology has to be taken into account: the irregular shaped particles of the second phase have to be substituted by modelled particles, which may be treated

- physically in the sense of field effects as stray fields
- stereologically so that measured parameters from two-dimensional polished sections may be transformed mathematically into parameters valid three-dimensionally.

Spheroid inclusions are appropiated best for this purpose. By introducing them into the homogeneous material - as done in fig.2 - a stray field is influenced and superimposed on the original field. As a consequence of this derivation the model is fixed to spheroidal inclusions. For them, both - the original as well as the stray field - may be described quantitatively by the respective field equation. Superimposing the two fields by combining the field

energies additively, the effective field and field properties result /7/. It also follows from the derivation that the field alteration depends on

- the concentration
- the shape and
- the orientation

of the inclusions, which appear as the microstructural parameters in equation 1

$$1 - c_D = \frac{\phi_D - \phi_C}{\phi_D - \phi_M} \left[\frac{\phi_M}{\phi_C}\right]^R \left[\frac{\phi_C + \phi_D \left\{1 - \frac{1}{2F_D + \cos^2\alpha\ (1 - 3F_D)}\right\}}{\phi_M + \phi_D \left\{1 - \frac{1}{2F_D + \cos^2\alpha\ (1 - 3F_D)}\right\}}\right]^T \quad (1)$$

with

$$R = \frac{F_D\ (1 - 2F_D)}{1 - 2F_D - \cos^2\alpha_D (1 - 3F_D)}$$

$$T = \frac{F_D\ (1 - 2F_D)}{1 - 2F_D - \cos^2\alpha_D (1 - 3F_D)} + \frac{2F_D\ (1 - F_D)}{2F_D + \cos^2\alpha_D (1 - 3F_D)} - 1$$

governing the microstructural effects on the effective conductivity of a two-phase material (ϕ_C) which additionally depends on the conductivities of its phases (ϕ_M, ϕ_D). In this context the concentration factor (c_D) refers to the volume content of the included phase, the shape factor (F_D) is physically identical with the well known depolarization factor /12/ and the orientation factor ($\cos^2\alpha_D$) refers to the mean of the cosinus squares of the angles formed by the rotation axes of the spheroids and the direction of the field as shown in fig. 3 /7/.

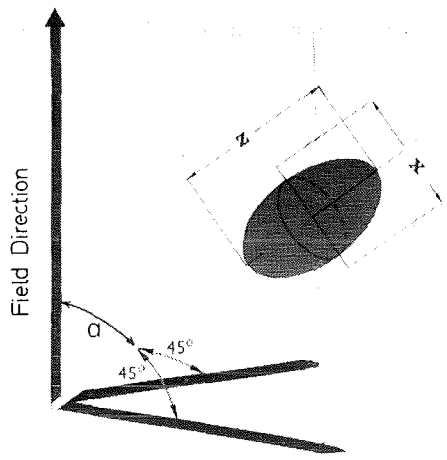

Fig.3: Orientation angle between field direction and spheroids rotation axis (z)

In the case where the ratio between the conductivities of the phases (ϕ_D/ϕ_M) becomes very small, the general equation simplifies to the following form

$$\phi_C = \phi_M(1-c_D)\,\frac{\cos^2\alpha_D - 1}{F_D - 1} + \frac{\cos^2\alpha_D}{2F_D} \qquad (2)$$

where again the three microstructural parameters appear, this time concerning the concentration, shape and orientation of pores. In fact, besides these three quantitative microstructural parameters, two other - qualitative - microstructural data govern the overall property being tacitly involved in the equations derivation, which are:

- the number of phases present in the material and
- the geometrical arrangement of the phases, which may correspond to a matrix or inteconnecting phase type microstructure.

Summarizing we have therefore to take into account five parameters to describe a material's microstructure sufficiently with respect to its field properties.

Fixing one of them in the general microstructure - property equation 1 - as for example the number of phases to be two- the equation's solution provides two values, forming property bounds versus concentration when plotted in a respective diagram as shown in fig. 4. These bounds are called first order bounds when based on the fixation of one - and only one - of the five defined microstructural parameters. As-

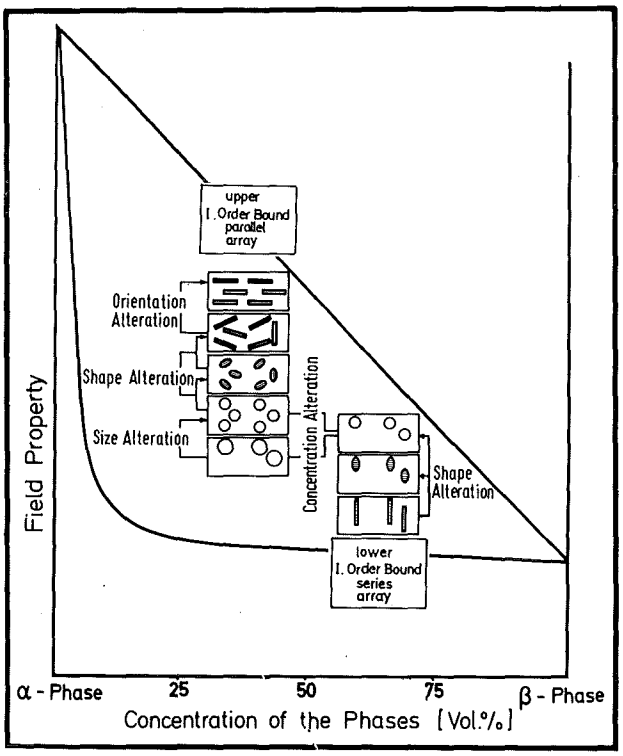

Fig. 4: Bound formation and property shifting schematically with respect to equation 1 and its underlying model microstructure

suming a two phase material, the first order bounds correspond to the utmost bounds given by Kirchhoff's law for parallel and series array of the phases. We get closer second order bounds /3/ by assuming a two phase isotropic material - fixed number of phases and fixed orientation - and third order bounds by supposing

- that the material is two phased
- that the material has an isotropic microstruc-

ture (orientation)
- that one phase forms the matrix.

Obviously the bounds become closer and therefore the property determination becomes increasingly more accurate with increasing information and accuracy about the material's microstructure, which may be obtained by quantitative microstructural analysis including stereology (compare fig.9). This is where and why stereology holds a key position in microstructure-property-correlations.

DETERMINATION OF MICROSTRUCTURAL PARAMETERS

As a consequence of the theoretical derivation the phase concentration factor in the equations refers to the volume content of one phase whilst - as mentioned above - the shape factor is identical with the depolarization factor when including spheroids in a homogeneous field /12/. As such it was derived as a function of the axial ratio $(\frac{Z}{X})$ of the spheroid as shown in fig. 5, where z is the rotation axis generally. The third microstructure parameter, the

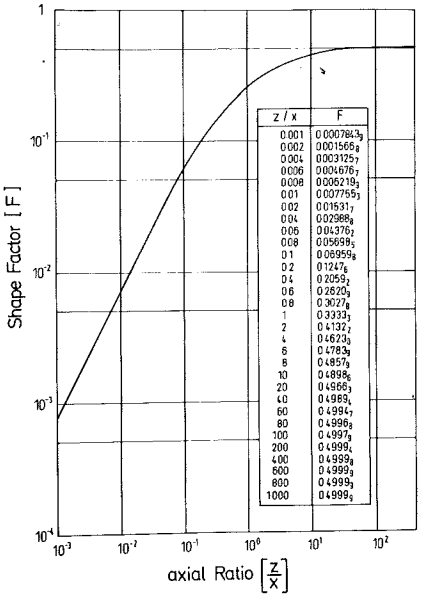

Fig. 5: The shape factor (F) as a function of the axial ratios $(\frac{Z}{X})$ of spheroids

orientation factor, also is a function of the axial ratio $(\frac{z}{x})$ and, - additionally - of the areal axial ratio $(\frac{a'}{b'})_A$ or $(\frac{b'}{a'})_A$ of ellipses measured in sections taken perpendicular to the field direction (compare also fig. 6):

$$\cos^2\alpha_D = \frac{[\frac{z}{x}]^2_{II}\,\overline{[\frac{b'}{a'}]^2_A} - 1}{[\frac{z}{x}]^2_{II}\,\{\overline{[\frac{b'}{a'}]^2_A} + \overline{[\frac{b'}{a'}]^2_B} + \overline{[\frac{b'}{a'}]^2_C}\} - 3} \qquad (3)$$

$$\cos^2\alpha_D = \frac{[\frac{z}{x}]^2_{=}\,\overline{[\frac{a'}{b'}]^2_A} - 1}{[\frac{z}{x}]^2_{=}\,\{\overline{[\frac{a'}{b'}]^2_A} + \overline{[\frac{a'}{b'}]^2_B} + \overline{[\frac{a'}{b'}]^2_C}\} - 3}$$

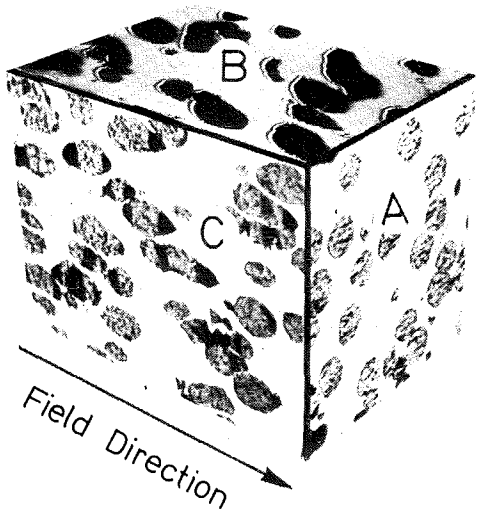

Fig. 6: Areal axial ratios of inclusions in sections taken parallel and perpendicular to the field direction

As can be seen from fig. 6 the difference between the areal axial ratios of sectioned ellipses in sections taken perpendicular to the field direction and those taken statistically through the material increases with increasing orientation.

The actual task now arising from the theoretically obtained definitions of the microstructural parameters is to calculate spatial - that means three dimensional - quantities from areal quantities measured in two-dimensional sections. This may be done by stereological relationships especially available for the case of spheroids, which was one crucial reason to prefer this model.

The areal axial ratios of a real material are determined as demonstrated in fig. 7 by measuring the area and the perimeter of the real features /11/. The adaption to the model is achieved substituting the real features by ellipses with a respective area to perimeter ratio.The transformation of their mean axial ratio into the one of the refering spheroid is solved stereologically and pointed out

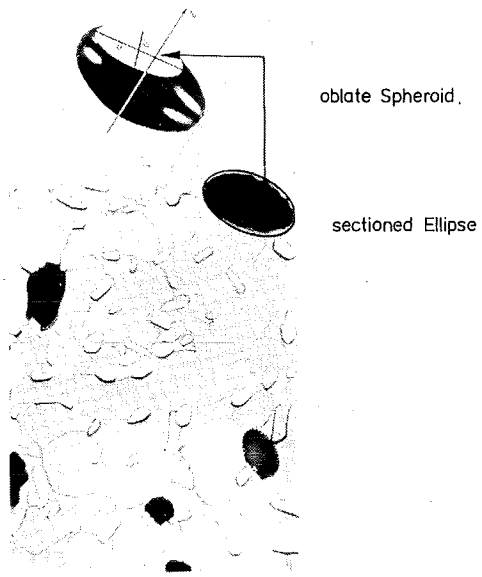

Fig. 7: The determination of substituting ellipses in cross sections

graphically in fig.8.Here one has to notice that one does not know whether the sectioned ellipses belong to an oblate or prolate spheroid. Therefore we have to take into account both which leads to two possible spatial axial ratios resulting in two shape factors and so returning to the bound concept; now, however, related directly to the real material and of higher order. - The explained determination

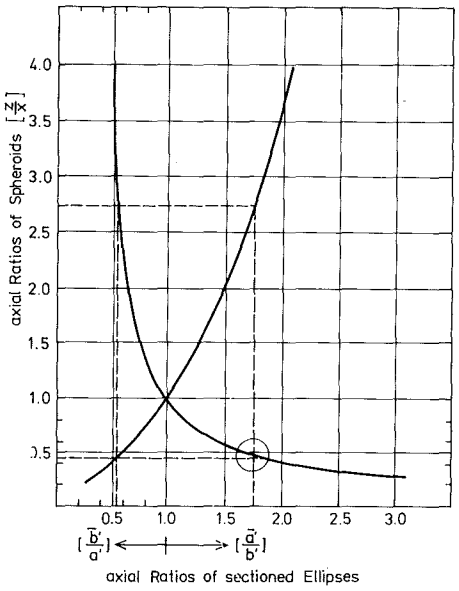

Fig. 8: The transformation of ellipsoidal to spheroidal axial ratios

of axial ratios also provides the orientation factor by measuring the areal ratios once in sections taken statistically through the material and another set taken perpendicular to the field direction (compare equ. 3 and fig. 6). So finally the phase concentration factor has to be determined, say the volume content of the inclusion which follows stereologically direct from the measured areal fraction due to the well known Delesse principle /13/.

By using model materials consisting of spheroidal pills as used in pharmacy embedded in a resin matrix phase, preparing cross sections of it, measuring and calculating their stereological quantities and comparing them with the known nominal quantities, the error for the relevant

quantities (axial ratios, volume content) was estimated to be about 5% (table 1) /9/.

Table 1: Comparison of nominal quantities with their corresponding values determined by quantitative microstructural analysis

	Quantity	Nominal	Measured	Rel. Err.(%)
Mean axial ratio in the plane	$(\overline{a'/b})$	1,2494	1,2027	3,88
	$(\overline{b/a'})$	0,8119	0,8541	5,19
Volume content	V_V	0,2180	0,2212	1,46
Mean axial length	$\overline{a'}$	6,0254	5,9103	1,94
Major axis	$a_=$	8,572	7,5254	13,90
	a''	8,231	8,0697	1,99
Minor axis	$b_=$	5,290	5,9819	11,32
	b''	6,190	6,2572	1,08
Rel. content of obl. and prol. spheroids	$N_=$	0,2121	0,1913	10,87
	$N_{\shortparallel}$	0,7879	0,8087	2,64
Mean axial ratio	$(\overline{z/x})$	1,1785	1,2001	1,83

In fig. 9 the steps necessary to determine the microstructural parameters with respect to microstructure-property-correlations are summarized. Quantitative microstructural analysis starts by statistical and selected sectioning of the material, which then has to be polished and etched to contrast the microstructural features /10/. By transforming their optical pictures to a monitor the contrasted phases can be measured electronically as done by automatic image analysers. The measured quantities form the input to a first stereological computer program, transforming the data from a two into the three dimensional state. A progressing stereological program provides the microstructural factors which fit into the microstructure-property-relationship, contained in a third computer program. This finally provides the properties of the material in dependence on its microstructure.

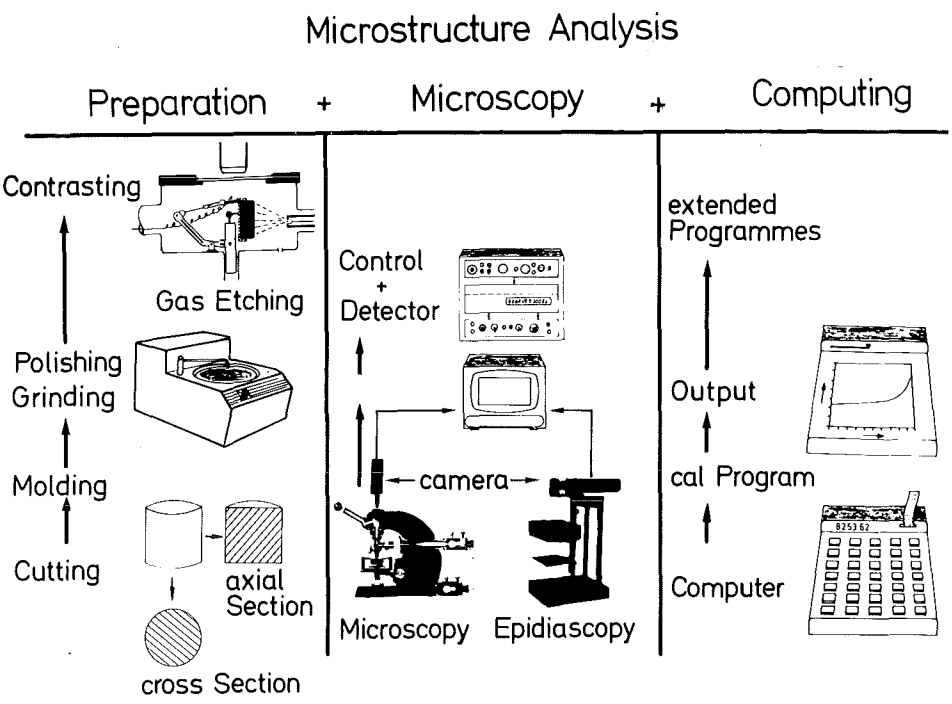

Fig. 9: Sequence of steps in quantitative microstructure analysis

COMPARISON OF MEASURED AND CALCULATED QUANTITIES

To check the reliability of the procedure and its underlying microstructural model in fig. 10 electrical conductivities of graphite-silver-composites have been compared with corresponding bounds. Due to its two-phase and isotropic but no more defined microstructure, second order bounds should hold for the electrical conductivity - as they do in the frame of an engineering approach. In the case of "Bamica" spheres, a special ceramic embedded in an alumina matrix phase, the dotted lines in fig. 11 should represent in the region and slope of the thermal conductivities, which - again - is sufficiently ful - filled by the experimental data. - In fig. 12 UO_2-almost spherical particles in different metal matrices scatter around the theoretical curve due to the deviations from speroidicity, but lucidly follow the calculated slope. - Finally, in fig. 13 the thermal conductivity of graphite fibre reinforced resin demonstrates plenty agreement between both the measured values and the calculated curve, which - together with additional comparisons /4,7/ - permits to

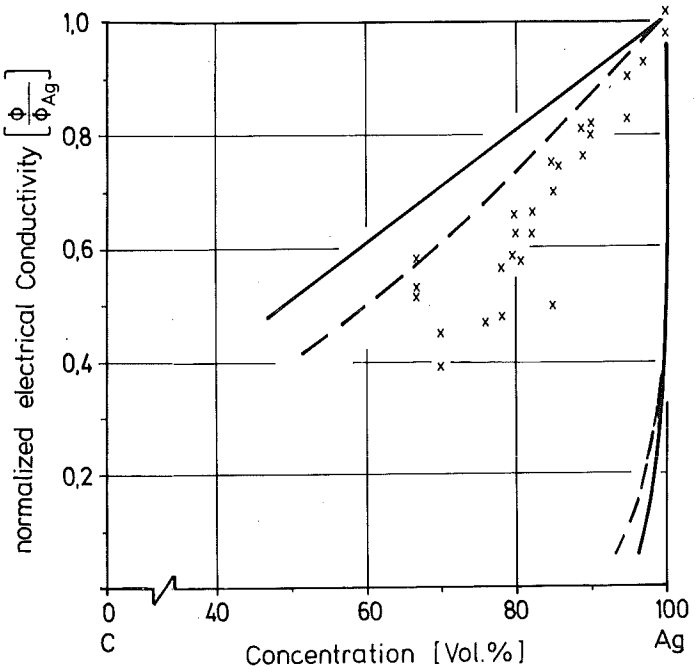

Fig.10: Electrical conductivities (x) of Ag-C-composites at room temperature and first (-) and second order bounds (---) /4/

conclude that in this context stereology is neither an impressive mathematical tool to play with, nor - as in the starting period of the discipline - mainly treated by "pure stereologists" - because of its handicap of resulting either in an inexact representation of real structures or of an exact representation of unreal models.

Stereology nowadays has to - and does - provide progress in materials science and practise and is - therefore - a key to the gates of tomorrow.

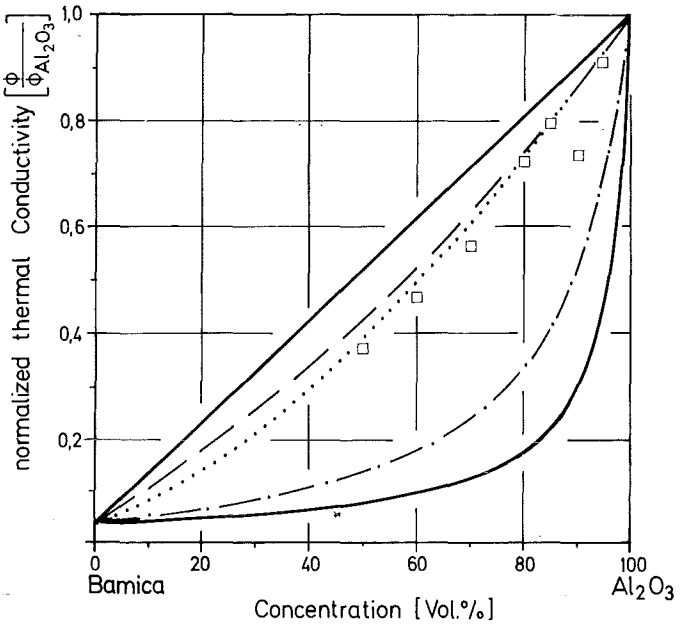

Fig.11: Thermal conductivities (x) of Bamica"-Al_2O_3-ceramics at 673 K and I. (-), II. (---) and III. order bounds (····) /4/

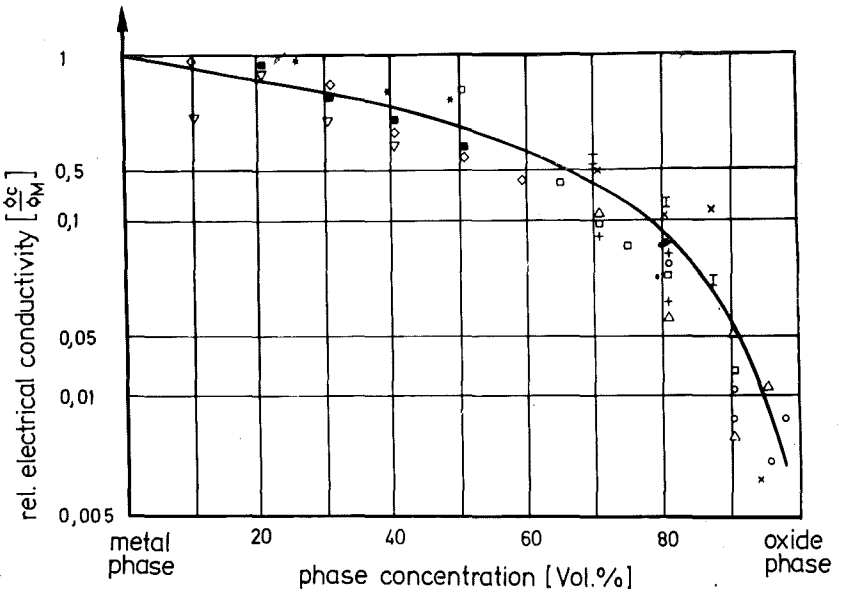

Fig.12: Electrical conductivities of UO_2-cermets with metal matrices and theoretical curve corresponding to spherical inclusions /8/

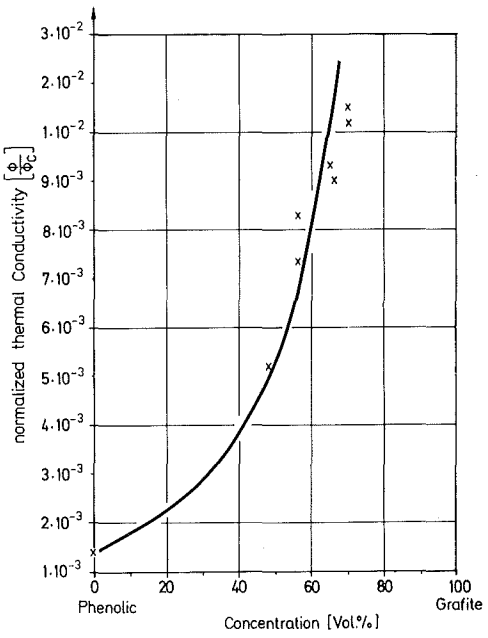

Fig. 13: Thermal conductivities (x) of graphite fiber-phenolic matrix phase - composites at room temperature and corresponding theoretical curve /4/

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Acknowledgement

Mrs. Karcher has carefully prepared the figures, Mrs. Nuss has written the text. The assistance of the two ladies is gratefully acknowledged by the author.