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Hardmetals — Metallographic determination of microstructure —

Part 2: Measurement of WC grain size

Métaux-durs — Détermination métallographique de la microstructure —

Partie 2: Mesurage de la taille des grains de WC

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Foreword

ISO (the International Organization for Standardization) is a worldwide federation of national standards bodies (ISO member bodies). The work of preparing International Standards is normally carried out through ISO technical committees. Each member body interested in a subject for which a technical committee has been established has the right to be represented on that committee. International organizations, governmental and non-governmental, in liaison with ISO, also take part in the work. ISO collaborates closely with the International Electrotechnical Commission (IEC) on all matters of electrotechnical standardization.

The procedures used to develop this document and those intended for its further maintenance are described in the ISO/IEC Directives, Part 1. In particular, the different approval criteria needed for the different types of ISO documents should be noted. This document was drafted in accordance with the editorial rules of the ISO/IEC Directives, Part 2 (see www.iso.org/directives).

Attention is drawn to the possibility that some of the elements of this document may be the subject of patent rights. ISO shall not be held responsible for identifying any or all such patent rights. Details of any patent rights identified during the development of the document will be in the Introduction and/or on the ISO list of patent declarations received (see www.iso.org/patents).

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For an explanation of the voluntary nature of standards, the meaning of ISO specific terms and expressions related to conformity assessment, as well as information about ISO's adherence to the World Trade Organization (WTO) principles in the Technical Barriers to Trade (TBT), see www.iso.org/iso/foreword.html.

This document was prepared by Technical Committee ISO/TC 119, *Powder metallurgy*, Subcommittee SC 4, *Sampling and testing methods for hardmetals*, in collaboration with the European Committee for Standardization (CEN) Technical Committee CEN/SS M11, *Powder metallurgy*, in accordance with the Agreement on technical cooperation between ISO and CEN (Vienna Agreement).

This second edition cancels and replaces the first edition (ISO 4499-2:2008), which has been technically revised.

The main changes compared to the previous edition are as follows:

— former 3.1 has been removed;

— 3.2 has been expanded;

— in Clause 5, “Electron back scatter diffraction (EBSD)” has been added;

— in 7.2.1, the list has been revised;

— in 7.3.3, Table 1, row “Electron back scatter diffraction” has been added and in the row “Scanning electron microscope”, the value for the “Minimum visible intercept length” has been corrected from 200 nm into 400 nm.

A list of all parts in the ISO 4499 series can be found on the ISO website.

Any feedback or questions on this document should be directed to the user’s national standards body. A complete listing of these bodies can be found at www.iso.org/members.html.

Hardmetals — Metallographic determination of microstructure —

Part 2: Measurement of WC grain size

1 Scope

This document gives guidelines for the measurement of hardmetal grain size by metallographic techniques only using optical or electron microscopy. It is intended for WC/Co hardmetals (also called cemented carbides or cermets) containing primarily tungsten carbide (WC[1)]) as the hard phase. It is also intended for measuring the grain size and distribution by the linear-intercept technique.

This document essentially covers four main topics:

— calibration of microscopes, to underpin the accuracy of measurements;

— linear analysis techniques, to acquire sufficient statistically meaningful data;

— analysis methods, to calculate representative average values;

— reporting, to comply with modern quality requirements.

This document is supported by a measurement case study to illustrate the recommended techniques (see Annex A).

This document is not intended for the following:

— measurements of size distribution;

— recommendations on shape measurements. Further research is needed before recommendations for shape measurement can be given.

Measurements of coercivity are sometimes used for grain-size measurement, however, this document is concerned only with a metallographic measurement method. It is also written for hardmetals and not for characterizing powders. However, the method can, in principle, be used for measuring the average size of powders that are suitably mounted and sectioned.

2 Normative references

The following documents are referred to in the text in such a way that some or all of their content constitutes requirements of this document. For dated references, only the edition cited applies. For undated references, the latest edition of the referenced document (including any amendments) applies.

ISO 3369, *Impermeable sintered metal materials and hardmetals — Determination of density*

ISO 3738-1, *Hardmetals — Rockwell hardness test (scale A) — Part 1: Test method*

ISO 3738-2, *Hardmetals — Rockwell hardness test (scale A) — Part 2: Preparation and calibration of standard test blocks*

ISO 4489:2019, *Hardmetals — Sampling and testing*

1) DE: Wolframcarbid, EN: tungsten carbide.

ISO 6507-1, *Metallic materials — Vickers hardness test — Part 1: Test method*

ISO 6507-2, *Metallic materials — Vickers hardness test — Part 2: Verification and calibration of testing machines*

ISO 6507-3, *Metallic materials — Vickers hardness test — Part 3: Calibration of reference blocks*

ISO 6507-4, *Metallic materials — Vickers hardness test — Part 4: Tables of hardness values*

3 Terms, definitions, symbols and abbreviated terms

3.1 Terms and definitions

For the purposes of this document, the following terms and definitions apply.

ISO and IEC maintain terminological databases for use in standardization at the following addresses:

— ISO Online browsing platform: available at https://www.iso.org/obp

— IEC Electropedia: available at http://www.electropedia.org/

3.1.1
nano
with WC grain size <0,2 µm

Note 1 to entry: Measured by the mean-linear-intercept method described in this document.

3.1.2
ultrafine
with WC grain size 0,2 µm to 0,5 µm

Note 1 to entry: Measured by the mean-linear-intercept method described in this document.

3.1.3
submicron
with WC grain size 0,5 µm to 0,8 µm

Note 1 to entry: Measured by the mean-linear-intercept method described in this document.

3.1.4
fine
with WC grain size 0,8 µm to 1,3 µm

Note 1 to entry: Measured by the mean-linear-intercept method described in this document.

3.1.5
medium
with WC grain size 1,3 µm to 2,5 µm

Note 1 to entry: Measured by the mean-linear-intercept method described in this document.

3.1.6
coarse
with WC grain size 2,5 µm to 6,0 µm

Note 1 to entry: Measured by the mean-linear-intercept method described in this document.

3.1.7
extra coarse
with WC grain size >6,0 µm

Note 1 to entry: Measured by the mean-linear-intercept method described in document.

3.2 Symbols and abbreviated terms

For the purposes of this document, the following symbols, abbreviations and units apply.

A	is the area, in square millimetres (mm^2)
d_{wc}	is the arithmetic mean linear intercept of WC grains, in micrometres (µm)
ECD	is the equivalent circle diameter, in millimetres (mm)
L	is the line length, in millimetres (mm)
LI	is the arithmetic mean-linear-intercept distance, in micrometres (µm)
l_i	is the measured length of individual intercepts, in micrometres (µm)
$\sum l_i$	is the sum of the measured length of each individual intercept
N	is the number of grain boundaries traversed
n	is the number of WC grains intercepted
m	is the magnification
m_{max}	is the maximum magnification
m_{min}	is the minimum magnification
s_m	is the measured size, in millimetres (mm)
s_a	is the actual size, in millimetres (mm)
EBSD	is electron back scatter diffraction
SEM	is scanning electron microscopy
FESEM	is field emission SEM
TEM	is transmission electron microscopy
LOM	is low magnification

4 General information

This document addresses the issue of good practice for the measurement of a mean value for WC grain size. It recommends the use of a linear-intercept technique for obtaining data. The measurements shall be made using good practice for the preparation of suitable microstructures for examination outlined in ISO 4499-1.

The properties and performance of hardmetals are directly dependent on the microstructure developed during manufacture, which in turn is controlled by the character of the starting powder batch. Understanding the microstructure is the key to controlling or improving properties, and therefore the measurement of microstructural features, particularly grain size and size distribution, is of paramount importance.

Methods of metallographic preparation and etching techniques are as important as the grain-size measurement method (see References [1] to [6]), and are included in ISO 4499-1. The principal type of hardmetal considered is WC with a Co binder. However, the procedure can be used for hardmetals that contain cubic carbides or which are based on TiC or Ti(C, N).

The most direct way to measure 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 area counting or by linear-intercept techniques.

There are three ways by which the mean size by number of the WC grains can be defined:

— by length (of a line across a 2D section of a grain);

— by area (of 2D sections of grains);

— by volume (of individual grains).

A number average is obtained by counting each measurement of the parameter of interest (length, area or volume) and dividing the total value of the parameter (length, area or volume) by the number of this parameter counted.

The value most used to date has been a length parameter. This can be obtained in several ways, for example, by parallel lines or circles as described in ASTM E112[14]:

— by linear intercept, called the Heyn method, from a straight line drawn across the structure;

— by the equivalent circle diameter: this is obtained by measuring grain areas and then taking the diameter of a circle of equivalent area. It is possible, for equiaxed grains, to convert an equivalent circle diameter (ECD) grain size to a linear intercept (LI) value using Formula (1).

$$\mathrm{LI} = \sqrt{A} = \sqrt{\pi / 4}\ \mathrm{ECD} \qquad (1)$$

Thus ECD = 1,13 LI.

This expression is discussed in References [1] and [7].

An additional method is that established by Jefferies, where the number of grains per unit area can be counted. This can, if required, be converted to an equivalent circle diameter.

It shall be noted that

— point/area counting provides no information on distribution, and

— the Jefferies method is not intended for use on multiphase materials such as hardmetals.

The recommended technique for measurement of hardmetal grain size is the linear-intercept method.

5 Apparatus

Grain-size measurements are obtained from images of the microstructure. ISO 4499-1, ASTM B657[12] and ASTM B665[13] should be consulted for best practice in the preparation of surfaces for imaging.

Hardmetal structural images are usually generated by either optical microscopy, scanning electron microscopy (SEM) or electron back scatter diffraction (EBSD). For accurate measurements, it is better to use scanning electron-microscopic images. Even in coarse-grained materials, the imaged surface cuts through a substantial number of the corners of grains, giving a proportion of small intercepts that can only be measured accurately using the scanning electron microscope.

Measurements of intercept lengths from the acquired images can be obtained manually or semiautomatically using image analysis. Automatic image analysis can be used in some circumstances when the images are fairly coarse and good contrast can be obtained, but for many materials, especially those with very fine grain sizes, good images are difficult to acquire and are generally not amenable to automatic analysis.

For the ultrafine and nano grades, good images are particularly difficult to acquire using conventional scanning electron microscopes with tungsten-filament electron sources. For these materials, it is

recommended that a field emission SEM (FESEM) be used. These systems give significantly higher resolution images, sufficient to measure materials with mean intercept sizes of about 0,1 µm to 0,2 µm. For materials with ever smaller grain sizes, it might be necessary to use transmission electron microscopy (TEM). However, the problems of sampling and specimen preparation are particularly severe (see Reference [9]). Careful specimen preparation for good images is vital for these materials, and often a combination of etching methods is helpful (see ISO 4499-1).

6 Calibration

To give reliable quantitative measurements, images shall be calibrated against a stage micrometer or scale traceable to a national reference standard. The most commonly used stage micrometers for SEMs are the SIRA grids. These are ruled lines which form a grid and are available with 19,7 lines per mm and 2 160 lines per mm. However, these shall also be calibrated and certified as being traceable to a national reference standard.

For images obtained from an optical microscope, an image of the calibration graticule shall also be obtained using the same objectives (and internal magnification step changers or zoom position) and illuminating technique. The microscope shall be set up for Köhler illumination to obtain the maximum resolution (see Reference [10]).

For images obtained from a scanning electron microscope, images of the graticule should be obtained under the same conditions (accelerating kV, working distance, illumination aperture) as those used for the hardmetal.

7 Grain-size measurement by the linear-intercept method

7.1 General

It is recommended that the arithmetic mean-linear-intercept be used as the parameter to define WC grain size. This is the simplest procedure to use and has the added advantage of providing data that can be used to quantify distribution width.

This method requires a straight line to be drawn across a calibrated image. In a single-phase material the length of line (L), starting at a random position, traversing a number of grain boundaries (N), and ending at another random position, is measured. The mean-linear-intercept distance LI is specified in Formula (2):

$$\mathrm{LI} = L / N \tag{2}$$

As can be seen from the Formula (2), only the the mean-linear-intercept distance is calculated, there is no information obtained on grain-size distribution.

For a nominally two-phase material such as a hardmetal (α and β phase), the linear-intercept technique is less straightforward because each phase shall be measured independently, but it can provide information on grain-size distribution. A line is drawn across a calibrated image of the microstructure of a hardmetal. Where this line intercepts a grain of WC, the length of the line (l_i) is measured using a calibrated rule (where i = 1, 2, 3, ..., n, for the 1st, 2nd, 3rd, ..., nth grain). It is advisable to count at least 100 grains, preferably at least 200 grains in order to reduce the uncertainty to below 10 %.

The mean-linear-intercept grain size is defined as Formula (3):

$$d_{\mathrm{wc}} = \sum l_i / n \tag{3}$$

Hardmetal grain sizes generally fall in the range 0,1 µm to 10 µm. Because of the uncertainties of measurement, it is good practice to report the the mean-linear-intercept grain size to one decimal place for values > 1,0 µm and to two decimal places for values < 1,0 µm, i.e. the results are reported to two significant figures, such as 3,4 µm or 0,18 µm.

A worked example is given in Annex A.

7.2 Sampling

7.2.1 Sampling of products

Sampling is the procedure whereby an item of hardmetal or a region within an item is chosen for testing. Random sampling is defined such that, in selecting an individual from a population, each individual in the population has the same chance of being chosen (see Reference [11]).

ISO 4489:2019, Clause 4 states: "For confirmation of the grade of hardmetal, it is usually sufficient to take a test sample of one unit". The following tests shall be carried out in accordance with the International Standards given in ISO 4489:2019, 5.1:

— Determination of coercivity	no standard available;
— Determination of density	ISO 3369;
— Determination of Rockwell hardness HRA	ISO 3738-1 and ISO 3738-2;
— Determination of Vickers hardness HV	ISO 6507-1, ISO 6507-2, ISO 6507-3 and ISO 6507-4.

and tests which may be carried out in special cases:

— Determination of microstructure	ISO 4499 (all parts);
— Determination of porosity and uncombined carbon	ISO 4499-4.

7.2.2 Sampling of microstructure

Sampling for microstructural purposes should be carefully considered depending on the reason for undertaking the measurements.

a) General check measurement of a sectioned isolated object

   — The images chosen for analysis should be representative of the whole section and should be obtained by random positioning. The number of images to be prepared is recommended to be at least four, which can be intensively analysed so that in total, at least 200 grains are measured.

b) Determination of homogeneity of grain size

   — In this case, a systematic set of images from defined locations within the section shall be obtained and intensively analysed so that at least 200 grains are measured from each location. This allows for example, trends in grain size greater than the likely error of measurement at each position (fractional error is proportional to $1/\sqrt{N}$, where N is the number of grains at each location) to be determined.

c) Inhomogeneous materials

   — In cases where the microstructure is inhomogeneous from one field of view to the next, it is good practice to increase the number of images evaluated, but to evaluate them less intensively, while still achieving a total feature count of > 200.

The magnification of the image obtained should be such that there are between 10 WC and 20 WC grains across the field of view, permitting individual intercepts to be measured to better than 10 % accuracy. This will usually allow 3 or 4 linear-intercept lines to be drawn across the image without intercepting any individual WC grain more than once. Most hardmetals have little or no anisotropy of structure, so it is unimportant if more or less parallel lines are used. If anisotropy is suspected, then it is better to

orientate the lines randomly and permit their intersection (see Reference [13]). Thus, from each image, about 50 linear grain-size intercepts may be obtained.

7.3 Measurement errors

7.3.1 Systematic and random errors

Measurement errors can have several sources:

— systematic, such as during calibration of the microscope;

— experimental or accidental, such as during data transfer or calculation of actual intercept lengths;

— statistical, such as due to the random nature of the microstructure.

A possible cause of a systematic error is that of calibrating the image from which measurements are to be made. In general, a single number is obtained for the magnification of an optical microscope. But, if the calibration is over different lengths or by different operators, the results will vary, producing a mean magnification and associated standard deviation. Errors are likely to be larger when using an SEM owing to magnifications not being fixed steps.

Accidental or personal errors occurs when measuring individual linear intercepts of WC grains. Different operators measuring along the same intercept line will not choose exactly the same intercept positions or might not detect all boundaries, and this leads to an uncertainty in the measurement. Accidental or personal errors are more difficult to quantify than systematic errors.

Statistical errors can arise if microstructures are inadequately sampled, for example, too few micrographs are used, or too few grains are measured. A useful test of the adequacy of the statistics is to perform a running-average test. As measurements are made, the mean-linear-intercept size or other parameters are continually re-computed to give running averages which are plotted against the total number of measurements made. The mean result will be seen to fluctuate but to converge towards the true mean for the microstructure with increasing numbers of measurements. Measurements can be halted when the residual fluctuations in the mean are adequately small.

7.3.2 Large WC grain sizes

Before deciding which magnification to use for grain-size measurement, a preliminary scan of the etched surface is useful to determine if there are large grains of WC present. If too high a magnification is used, these grains may not fit into the field of view and this will thus affect the measurement statistics. Ideally, the magnification used should be such that the largest WC grain imaged is at most (giving 10 to 20 grains across the view) a third of the field of view as a general guide. In practice, there will always be large grains that intercept the edges of the field of view and thus are not measured. However, if sufficient fields of view are measured and the running-average technique (see Figure A.4) is used, the affect of large grains is minimised.

7.3.3 Smallest measurable intercept

At the time of publication, there are no standards available relating to the smallest intercept size that can be measured by either optical or scanning electron microscopy. As a guide, the resolution of the instrument used can be used to determine the smallest intercept which can be measured. For a particular resolution, the lower limits to which linear intercepts can be measured are given in Table 1. These figures represent the highest resolution which can be obtained under optimum conditions. In practice, poorer resolution can be obtained, particularly when it is difficult to obtain high resolution images because of problems with surface preparation. Thus the smallest intercept which can be sensibly measured will increase. In practice, the smallest intercept measurable is twice the resolution of the instrument and the uncertainty of the measurement is twice the resolution.