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NEUTRON DIFFRACTION TEXTURE ANALYSIS OF MULTIPHASE AND LOW-SYMMETRY MATERIALS USING THE POSITION-SENSITIVE DETECTOR JULIOS AND PEAK DECONVOLUTION METHODS

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ABSTRACT --- Diffraction patterns of multiphase and low-symmetry materials are characterized by a manifold of reflections with high peak overlap, hindering individual pole figure measurements by conventional X-ray technique. These difficulties can be overcome by using neutron diffraction with its advanced position-sensitive detector technology and by separating overlapping peaks by means of profile fitting methods. The paper describes appropriate instrumental equipment and a semi-automatic procedure for individual pole figure data analysis to be performed interactively on a PC. Pole figure results of a two-phase chalcopyrite ore are given.

INTRODUCTION

X-ray and neutron diffraction are well established techniques for texture investigations of polycrystalline material. Quantitative texture analysis is based on the measurement of preferred orientations of different crystallographic directions through their individual pole figures hkl and by combining those experimental pole figures mathematically to a three dimensional orientation distribution function (ODF).

In metallurgy, pole figure measurements and ODF calculations have been developed to a high standard and are widely performed on a routine basis. On the one hand, metal textures can be measured by X-ray techniques being easily available in many laboratories, and on the other hand, most metals are of high crystallographic symmetry, i.e. mostly cubic or hexagonal, strongly simplifying pole figure measurements of several individual hkl and the ODF calculation procedure.

The situation in geological texture analysis is quite different. Very often, X-ray diffraction, which measures the surface texture of a very small area of the specimen, is not the adequate method. This holds especially for coarse grained material and multiphase specimens. Here neutron diffraction is the favourite tool with the

disadvantage of its rather limited availibility. Most of the mineralogical constituents in geological specimens, like rocks or ores, are of low crystallographic symmetry, i.e. monoclinic or triclinic. Therefore, the diffraction patterns are characterized by a manifold of reflections originating as well from the individual mineral constituents as from the multiphase composition of the samples. Three dimensional texture analysis of low symmetry and multiphase specimens requires a large number of individual pole figures.

The measurement of individual pole figures, however, is complicated by peak overlaps ocurring in diffraction patterns of low symmetry or multiphase materials. To overcome these difficulties, total peak profiles must be registered in order to have a chance of deconvoluting overlapping peaks by peak profile analysis methods and to obtain reliable pole figure data. Position sensitive detectors (PSDs) are the proper experimental means to study such samples. PSD installations and the application of PSD techniques are more advanced and less problematic in neutron diffraction than in X-ray diffraction.

The scope of this paper is to expose the special characteristics of neutron diffraction in context with pole figure measurements of multiphase and low-symmetry materials and to discuss the necessary instrumental conditions and analytical procedures.

NEUTRON DIFFRACTION CHARACTERISTICS

The fundamental aspect of using neutron diffraction in texture analysis is the high penetration capability of neutrons through matter. The absorption coefficients of neutrons for most materials are orders of magnitude smaller than those for X-rays [1]. Low absorption diminishes essential restrictions for pole figure measurements with respect to sample constitution, shape and size and with respect to diffraction geometry.

Due to the different absorption characteristics, X-ray and neutron diffraction are qualitatively different. The local or surface texture analysed by X-rays contrasts the global or volume texture investigated by neutrons. In geology, global textures are needed for the study of correlations between crystallographic preferred orientation and the macroscopic anisotropy of physical properties. Local inhomogenities often existing in natural material, roughness or surface effects from mechanical treatment are averaged in specimens of several cm^3 volume. Because of large sample volumes and neutron beam cross sections of about 100 x 50 mm^2 texture investigations of samples with grain sizes up to several mm can be performed with statistical significance.

In neutron diffraction, complete pole figures can be measured in transmission geometry. Using the sperical sample method, the whole sample is penetrated by neutrons and no absorption corrections have to be applied. The shape of the sample can vary from a sphere to a cube or a cylinder, thus simplifying the sample preparation. Pole figures can be collected using position sensitive detectors without the need of complicated intensity corrections, which arise in X-ray PSD diffraction technique through severe geometrical aberrations. In X-ray diffraction, transmission and reflection geometry of flat specimens is used requiring different intensity corrections due to absorption and defocusing effects.

INSTRUMENTAL ASPECTS

The Bonn University four-circle diffractometer, installed at a thermal beam tube of the FRJ-2 reactor in the Forschungszentrum Jülich has been designed to meet the special requirements of texture investigations. The wavelength used for standard operating conditions is 1.20 Å. The diffractometer can be operated alternatively with two different Eulerian cradles: a commercial one having an excentric phi-circle for installations of various sample arrangements and a special one installed completely inside the cooling chamber of a helium cryostat for variable temperature pole figure measurements [2]. Pole figure scans are measured in an approximated equal-area scan mode [3] with preselectable step widths for azimuth ϕ and pole distance χ. In the standard scanning schedule, data points are collected in about 800 different orientations on one hemisphere.

An essential component of the diffractometer setup is the detector installation. In conventional diffraction technique, which is still widely applied in X-ray texture diffractometry, the diffractometer is equipped with a single counting tube. It is the classical procedure to position the single channel counter into a hkl peak maximum and to register the orientation dependent intensities of one special crystallographic direction. Several pole figures have to be scanned one after another. The total number of possible pole figures is limited by the number of non-overlapped reflections in the diffraction pattern. In case of overlapping peaks the peak maximum intensity may be composed of several hkl contributions, which cannot not be separated, and this may give rise to serious errors.

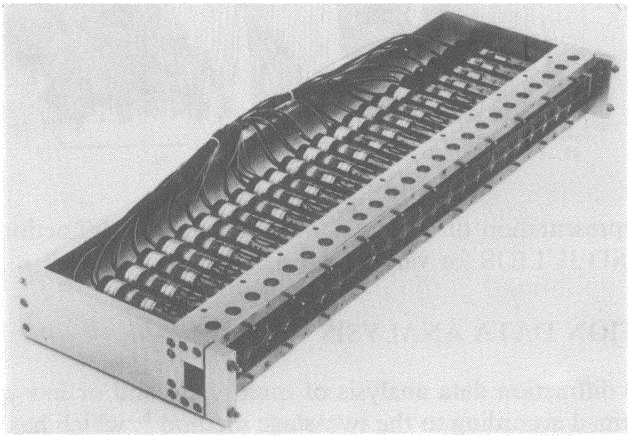

Fig.1: Scintillation detector JULIOS, the first linear position sensitive solid state detector installed at a neutron diffractometer

A dedicated neutron texture diffractometer is operated by using PSD techniques. Instead of measuring individual pole figures only in peak maximum positions, the PSD registers a large section of the diffraction diagram containing total peak profiles of many different hkl simultaneously, in our cases 20 to 30. The diffractometer of Bonn University is equipped with the scintillation PSD JULIOS (Fig. 1) [4], which has been specially developed for the requirements of pole figure data collection. The detector is of plane shape to allow variable distances between

sample and detector and thus variable angular resolution at the diffractometer. The sensitive length of JULIOS is 682 mm, corresponding to an angular range of about 35° in 2Theta in 1 m distance to the sample (Fig. 2).

The absolute length has been chosen to minimalize the so-called "blind area" during pole figure scanning [5]. The blind area arises for those pole figures, which are registered under non-bisecting scattering conditions at the outer parts of the PSD. In the case of JULIOS, the blind area is filled with only one additional data point in sample orientations $\chi = 90°$ and $\phi = 0°$ for each hkl. There is no need for additional scanning patterns in different sample adjustments (see also [3]).

The neutron data registered by the PSD JULIOS are stored in 1024 channels; each channel represents a 2Theta width of about 0.03°, when the detector is positioned in 1 m distance to the sample. The data point density has been adapted to the requirements of peak profile analysis. The reliability of the deconvolution of overlapped reflections by mathematical methods depends on the angular resolution of the diffractometer and on the data point density of the diffraction pattern (see [4]).

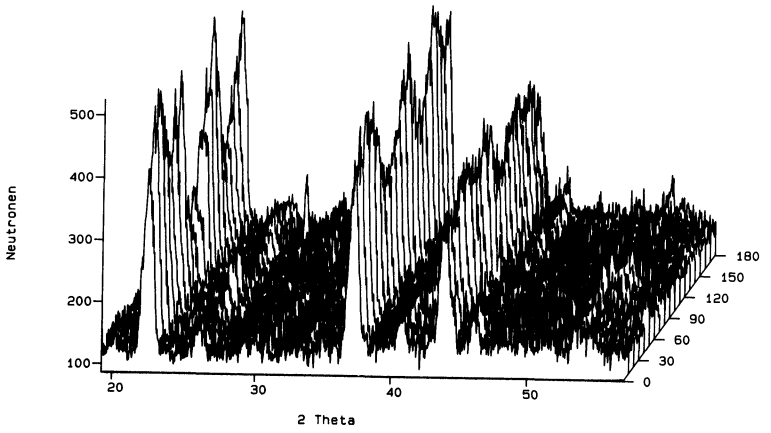

Fig. 2: 3d-representation of a sequence of chalcopyrite diffraction patterns registered with PSD JULIOS for varying ϕ-orientations.

DIFFRACTION DATA ANALYSIS

The PSD diffraction data analysis of multiphase and/or low-symmetry materials is performed according to the two-stage method [6], which has been developed in our institute for structure and texture analysis in X-ray and neutron powder diffractometry. Stage (1): Individual reflection data like position, halfwidth and integrated intensity are analysed applying mathematical procedures of profile fitting. Stage (2): The orientation dependent reflection intensities are processed via standard routines for pole figure representation.

The crucial point in the data analysis is the separation of overlapping peaks to be performed in stage (1), i.e. the pattern decomposition. This is done by fitting the diffraction patterns using the Gaussian profile function to describe the individual neutron peak profile [7]. The diffraction data profile analysis is performed on a two step basis.

The first step is concerned with the evaluation of global parameters, which are valid for all sample orientations, i.e. independent of the texture; global parameters are positions and halfwidths of the Bragg reflections. The determination of the global parameters is performed on a sum-data set, which is generated by adding up the individual diffraction patterns of all sample orientations. This sum-data set is characterized by very good statistics; it represents the scattered intensity from the sample during the total data collection time.

The second step is concerned with the determination of the texture specific parameters; these are the reflection intensities for the individual sample orientations. The intensity analysis has to be performed on the individual diffraction patterns, based on the refined global parameters of the sum-data set. By this procedure, the relatively low statistics of the individual diffraction patterns can be overcome.

We have developed a semi-automatic procedure (see [8]) for data evaluation and profile fitting of the about 800 diffraction patterns, which are collected during one pole figure scanning. The successive steps are:

1. generating a sum data set by adding up the 800 individual diffraction patterns,
2. interactive determination of a background function for the sum data set by using polygons or splines,
3. interactive determination of positions and halfwidths of all detected reflections by applying least squares based profile fitting on the sum data set (Fig. 3),
4. starting a batch job for the intensity analysis of the individual diffraction patterns performing
   a) adjustment of the sum-data set background function to the individual data sets by linear scaling,
   b) subtraction of the actual background and
   c) least squares intensity adjustment based on known peak position and halfwidth parameters.

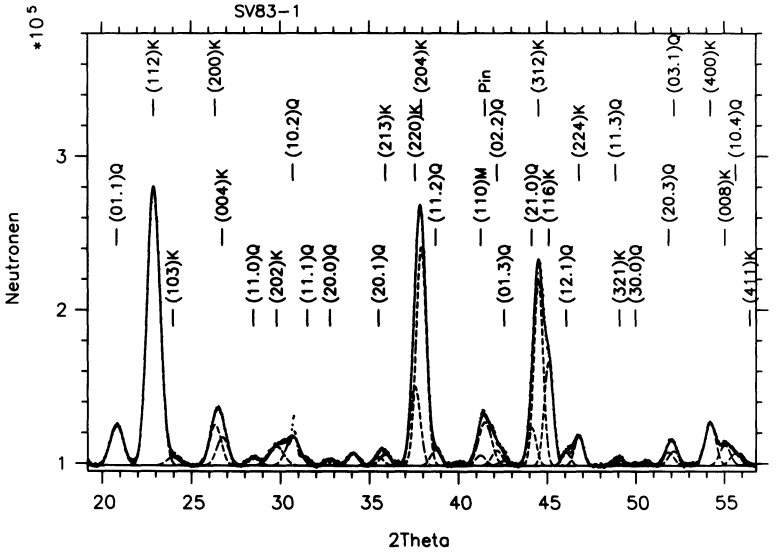

Fig. 3: Profile analysed sum diagram of a two-phase chalcopyrite ore. The indexing refers to the chalcopyrite (K) and quartz (Q) structure.

The data analysis routines, written in Turbo Pascal 5.5, are run on an IBM compatible AT personal computer. The mean computational time for the intensity analysis of about 800 diffraction patterns containing about 30 individual reflections (= pole figures) is about 2h using a 386 PC processor, or 6h using a 286 PC processor. The program package is based on the PC-program PROFAN-PC for individual powder peak profile analysis [9]. This program has been specially designed for on-line operation on a graphic display..

POLE FIGURE RESULTS

The University Bonn neutron texture diffractometer is operated as a service instrument, which is used for texture investigations in both material and earth sciences. Recently, the data analysis procedure descibed above has been applied on the texture analysis of naturally and experimentally deformed ores. Pole figure results of a two phase sample consisting of hematite and biotite, a mica with monoclinic symmetry, have been published [10].

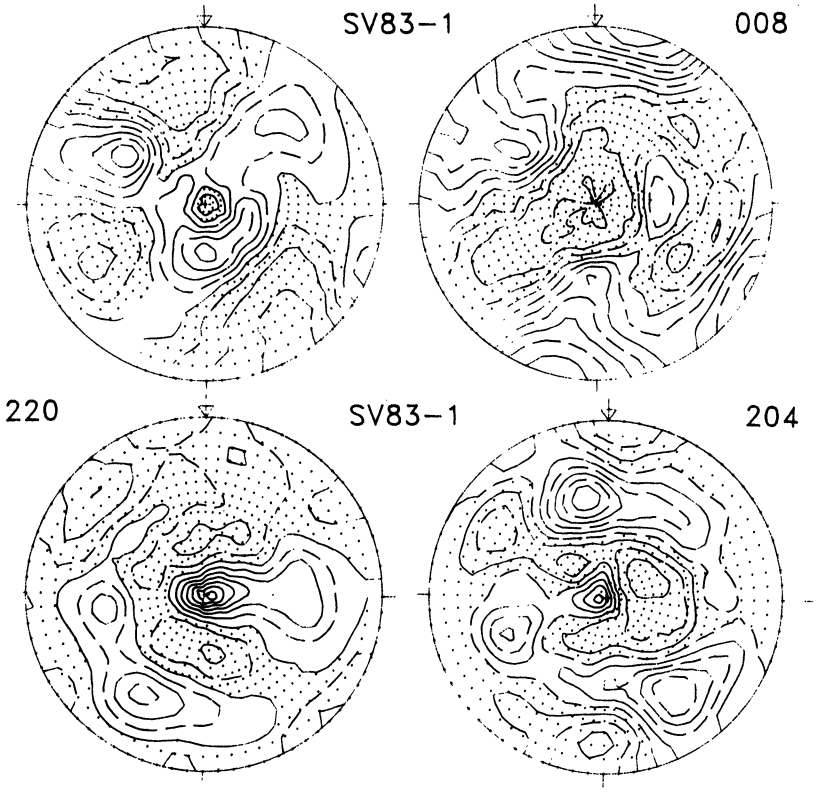

Fig. 4: Resolved chalcopyrite pole figures 400/008 (above) and 220/204 (below). Maximum and minimum pole densities are 1.97 and 0.30 for 400, 3.04 and 0.01 for 008, 2.67 and 0.24 for 220, and 1.57 and 0.62 for 204. Dotted sections represent pole densities < 1 mrd.

Here we present pole figure results of a two phase chalcopyrite ore consisting of about 70% chalcopyrite $CuFeS_2$ and 30% quartzite. Performing X-ray texture diffractometry, tetragonal close adjacent chalcopyrite reflections are not distinguishable from cubic reflections of the similar sphalerite structure, because of an unfavourable c/a-ratio of 1.97. For the study of pseudo-cubic or true tetragonal deformation behaviour of chalcopyrite, several natural and experimentally deformed samples have been analysed by neutron diffraction. A number of 32 reflections simultaneously recorded with the PSD installation described above have been profile analysed (see Fig. 3); 22 of them were considered to have large enough intensities for pole figure representation. Tetragonal overlapping reflections {h00}/{002h} and {hh0{/{h02h} have been separated. Some selected pole figures of the chalcopyrite phase are plotted in Fig. 4. The measuring time of one sample was about 50h, the computational time for the data analysis was about 6h. The interpretation of the preferred orientation behaviour of the chalcopyrite ore is given in this volume in a separate paper [11].

CONCLUSIONS

Neutron diffractometry in connection with a PSD is a most powerful technique in texture analysis. The simultaneous recording of many pole figures by the PSD technique yields reasonable measuring times. The analytical procedure of pattern deconvolution by profile fitting methods opens new possibilities in texture investigations of geological material.

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