Focusing High-Resolution Three-Axis Neutron Diffractometer for Investigations of Special Tasks of Powder Diffractometry

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Abstract. Feasibility of focusing high-resolution three-axis diffractometer with the polycrystalline sample between the monochromator and the analyzer was tested for studies of finer effects of powder diffraction lines and for investigations of special tasks of powder diffractometry. The focusing 3-axis diffractometer set-up equipped with bent perfect crystal monochromator and analyzer offers the sensitivity in determination e.g. of strains in polycrystalline materials **=*d*/*d* close to 10-5. Together with special tasks of strain/stress studies related namely, to plastic deformation, it permits to study a finer substructure of individual diffraction lines which can appear e.g. in case of polycrystalline alloys where more phases having very close values of the lattice spacing could exist. Resolution properties of the high-resolution diffractometer setting will be documented on several experimental results obtained on unconventional bulk samples.

1. Introduction

Neutron (as well as X-ray) powder diffraction is a wide spread method for determination of the structure and structural properties of polycrystalline materials. Conventional powder diffraction instrument is based on a two-axis diffractometer including a sophisticated monochromator, diffraction optics elements and a position sensitive detector (PSD) [1-4]. However, in some cases its *d/d* resolution suffer limitations coming e.g. from a necessity of using samples of small dimensions, a requirement of a large monochromator take-off angle and a high spatial resolution of PSD, which can be solved acceptable at high-flux neutron sources. In the special cases of elastic and plastic deformation studies of bulk polycrystalline samples when the whole powder diffraction spectrum is not required and/or identification diffraction peaks related to phases having very close lattice spacings, a high-resolution three-axis alternative can be helpful [5,6]. The alternative consists of an unconventional three axis set-up employing optimally bent perfect crystal (BPC) monochromator and analyzer with a polycrystalline sample in between. The alternative provides a substantially higher resolution and though it is much more time consuming than the conventional powder diffractometer it can be used even at the medium power neutron source. The aim of this paper is to prove a feasibility of using the alternative diffractometer setting for studying special material research tasks e.g. determination and volume content of different material phases having very close values of lattice spacing, or microstructure analysis of plastically deformed samples on the basis of diffraction profile analysis etc. [7-9].

1. Basic description of the 3-axis diffractometer set-up

As schematically drawn in Fig. 1, the testing neutron optics diffractometer installed at the research reactor LVR-15 in Řež and operating at the constant neutron wavelength of 0.162 nm was used for the feasibility studies. The diffractometer is equipped with the BPC Si(111) monochromator of the dimensions of 200 x 40 x 4 mm3 (length x height x thickness) and with the fixed radius of curvature *R*M of about 12 m. Ge(311) BPC analyzer was in the form of a rather thin slab of the dimensions of 200 x 40 x 1.3 mm3 which permitted us to bend it by a four point bending device in a large range of curvatures. The monochromator - sample distance, *L*MS, being 1.7 m and the sample - analyzer distance, *L*SA, being 0.5 m were fixed. The diffractometer operates in the open beam mode without any Soller collimators.



**Fig. 1.** Scheme of the tested 3-axis diffractometer settings using the BPC monochromator and the rocking BPC analyzer (*R*M, *R*A - radii of curvature, **M, **A - Bragg angles) for vertical – (a) and horizontal – (b) positions of a polycrystalline sample.

1. Resolution of the 3-axis set-up for bulk samples

During the extensive experimental studies several well annealed -Fe standard polycrystalline samples of the cylindrical form and 4 bent BPC analyzers (Ge(111), Ge(311), Si(220) and Si(400)) were used for investigation of basic resolution characteristics for -Fe(110) reflection and the detailed resolution properties can be found in ref. 6 and 10. Following the drawing shown in Fig. 1a the samples of several diameters of from 2 mm to 8 mm were situated in the vertical position. It is clear that the best resolution of the setting can be obtained when minimizing dispersion of the whole system. When solving the problem in momentum space, it means that the orientation of the *Δk* momentum elements related to the monochromator and analyzer should be matched to that of the sample, and this can be

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**Fig. 2.** Analyzer rocking curve for 8 mm -Fe(110) standard sample and the optimum radius of curvature of the analyzer of 9 m – (a), its *FWHM* dependence – (b) and the peak intensity – (c) on the analyzer curvature. The inserted points correspond to the other diameters of the sample.

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**Fig. 3.** Analyzer rocking curve related to 5 mm -Fe(110) standard sample situated at the horizontal position for the optimum radius of curvature of the analyzer of 9 m and for two widths of Cd slits: (a) – 10 mm and (b) – 20 mm.

achieved by proper optimum radii of curvatures of the bent perfect crystal monochromator and analyzer [5]. Then, it results in a high-resolution (a minimum *FWHM* of the analyzer rocking curve) and a maximum detector signal, simultaneously.Fig. 2 shows the best basic resolution characteristics as dependent on the curvature of the Ge(311) BPC analyzer. Inspection of Fig. 2 reveals that as a result of a simultaneous impact of the focusing in real and momentum space the best resolution of the setting as well as a maximum intensity was found for the same radius of curvature of the analyzer of *R*A=9 m. Furthermore, it has been found, that much larger widths (up to 20 mm) of the irradiated gauge volumes can be investigated by the three-axis setting when contrary to the conventional diffractometry, the resolution is much less affected (see Fig. 3). The width of the samples was simulated by situation of the samples in the horizontal position (see Fig. 1b) in combination with different widths of the Cd slits before the sample. In this case the contribution of the vertical dimension to the resolution was neglected. It can be seen from Fig. 3 that even for 20 mm width of the sample, the resolution represented by *FWHM* of the rocking curve is still good.

**4 Few possible applications**

First, we used several samples of the 3D printed material of gamma titanium aluminides (Ti48Al2Cr2Nb) loaded on the bulk Ti substrate. A powder fraction 45 – 150 µm was used. Manufacture of specimens was carried out by using the DED (Directed Energy Deposition) printing device InssTek MX-LAB. Laser power, speed of the printing and the dwell time were 130 W, 849 mm/min and 3.5 sec, respectively. Parameters related to the samples were: Both S1 (a preheat of the platform on 500 oC) and S2 (without a preheat) used cladding head SDM 800 and the 500 m hatch distance for the 800m spot size and the 250 m height of the layer. The following analyzer rocking curves (see Fig. 4) show how the differences in preparation of the samples reflect their *FWHM*. As to this sample it should be noticed that TiAl powder sample has not appeared to be a good scatterer for neutron diffraction.

 

**Fig**. **4.** Analyzer rocking curves related to the individual TiAl samples the **= 9 mm diameter and 20 mm height: (a) – powder sample, (b) – sample S1 and (c) – sample S2.



**Fig. 5.** Analyzer rocking curve re related to the wrought aged Inconel 718 sample.

Further experimental applications of this setting are demonstrated by using unconventional samples rarely used in conventional powder diffractometry. First -Fe(110) powder in 5 mm V container situated in a vertical and horizontal position was used (see Fig. 5). It can be seen from Fig. 5 that even for a sample 20 wide

(simulated by the Cd slit) the *FWHM*A of the analyzer rocking curve could be acceptable in some medium-resolution powder diffraction experiments.







**Fig.** **5.** Analyzer rocking curves related to the -Fe(110) powder in a Vanadium container of the 5 mm diameter situated on the sample position : (a) - vertically and (b) and (c) – horizontally.

For next samples, we used the steel cylindrical solid of the diameter of 18 mm (see Fig. 6), the steel tube with the external diameter of 16 mm and the thickness of the wall of 1.6 mm (see Fig. 7) set both in the vertical and horizontal position and -Fe plate of the thickness of 5.2 mm in the vertical position. Detailed inspection of the Fig. 6a reveals that the rocking curve is left-right asymmetrical (has no Gaussian form) due to the absorption of neutrons overcoming different path lengths in the material when looking along the axes of the incident and diffracted monochromatic beams. Similarly, in case of Fig. 7a the diffraction profile of the left part of the tube is slightly separated from the right one related to the irradiated right part when looking along the axis of the diffracted monochromatic beam. Therefore, the diffracted neutrons are impinging on spatially different parts of the bent analyzer. On the other hand, for samples horizontally situated in the setting the rocking curves are smooth and of course, their *FWHM*s depend of the width of the irradiated samples simulated by the width of the Cd slit. Furthermore, it has been observed that, as expected, in all cases with the large width of the irradiated sample (e.g. the case of the Cd slit of 20 mm) the rocking curves do not become Gaussian. However, for some types of experiments (e.g. when studying changes of the lattice spacing) it should not be a principal problem.





**Fig. 7.** Analyzer rocking curves related to the **= 16 mm diameter steel tube (thickness of the wall of 1.6 mm) situated: (a) – vertically and (b) - horizontally.





**Fig.** **8.** Analyzer rocking curves related to the -Fe(110) plate of the 5.2 mm thickness situated vertically for Cd slits of the width: (a) - 10 mm and (b) – 20 mm.







**Fig. 6.** Analyzer rocking curves related to the cylindrical steel solid of the 17 mm diameter situated: (a) – vertically and (b) and (c) – hori-zontally.

**5 Conclusion**

Resolution properties of the three-axis setting employing BPC Si(111) monochromator and BPC Ge(311) analyser with the -Fe(110) polycrystalline standard samples between them were tested on the NPI neutron optics diffractometer. Some basic resolution results are shown in Figs. 2 and 3. It is necessary to point out that the optimisation of the setting was limited by the fixed curvature of the installed Si(111) monochromator and its take-off angle as well as the mutual distances between the monochromator, the sample and the analyzer. Then, in some special cases of unconventional bulk samples (up to about 20 mm width) the feasibility of using such setting for investigations requiring higher resolution than conventional instruments is documented. The resolution of the setting for the Ge(311) analyzer shows a minimum in the vicinity of 1/*R*A = 0.11 m-1.

The resolution represented by *FWHM* of the analyzer rocking curve depends on many parameters [5] and for a design of the fully optimized three-axis setting which incorporate all free parameters, Monte Carlo simulation is recommended [11].

As expected, thanks to rather low scattering angle, the setting with -Fe(110) sample appears quite luminous with a high angular resolution *FWHM* because it exploits much more (at least partly) focusing in real and momentum space, simultaneously. In this case the lattice spacing of -Fe(110) is much closer to the one of the Ge(311) analyzer. The curvature 1/*R*A related to the best achieved resolution is situated within the range of the available analyzer curvatures. Therefore, some increase of the luminosity of the setting can be achieved by using a thicker crystal analyzer, e.g. up to 4 mm.

The following best values of *FWHM* obtained for different diameters of the standard sample were: 0.13 deg - **= 8 mm, 0.10 deg - **= 5 mm and 0.07 deg - **= 2 mm for Ge(311) analyzer.

Feasibility of the three-axis setting for some high-resolution studies on the bulk samples of rather large width simulated by different slit widths (see Fig. 3) was documented. I was shown that even for 20 mm slit width, the mildly relaxed resolution is still sufficient for e.g. possible application of the analysis of the position and shifts of the diffraction peak profiles [7-9]. In comparison with the conventional two-axis set-up, this alternative instrument could be successfully applied, namely, in the strain/stress measurements in bulk samples exposed to a thermo-mechanical external load, e.g. in tension/compression rig, in aging machine etc. It means in the cases when the whole powder diffraction spectrum is not required and the position of the sample is fixed. Thus, it can provide average strain/stress values over a large irradiated volume. Otherwise, for comparison of different samples attention should be paid to keeping the central point of the irradiated gauge volume in the same position with a high accuracy to avoid a shift error to **A. Naturally, the high-resolution setting can also be attractive for studies of a fine substructure of e.g. alloys which could hardly be seen by using the conventional powder diffractometer [12]. Of course, that the pre-sented setting can also be used for macro-strain scanning, but due to the analyzer step-by-step analysis, the measurement would be rather time consuming.

In conclusion, we hope that the presented neutron diffraction setting can offer an additional support and complement the information obtained using other methodologies.

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