OPTIMIZED STRAIN/STRESS DIFFRACTOMETER EQUIPPED WITH FOCUSING BENT PERFECT CRYSTAL MONOCHROMATOR WOULD PERMIT SOME KINETIC PROCESSES IN POLYCRYSTALLINE MATERIALS

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Abstract
In this paper properties of the dedicated neutron strain/stress diffractometer installed at the beam port ST-1 of HANARO reactor in KAERI are described. Thanks to the employment of the horizontally focusing bent perfect crystal monochromator and the optimization procedure the resolution and namely the luminosity of the instrument permit us to study kinetic processes in polycrystalline materials running within a few seconds.

Introduction

The neutron strain/stress scanner evaluates the variations of lattice spacing within a sample with a spatial resolution of the order of mm given by the dimensions of the gauge volume. Typical neutron diffractometer dedicated for strain/stress measurement at a reactor source is shown schematically in Fig. 1 [1,2]. The polychromatic neutron beam is first monochromated to a chosen wavelength by diffraction from a suitable monochromator. This monochromatic beam of a suitable size and divergence is given by the use of appropriate beam defining elements and is then diffracted from the specimen. In a similar way, the geometry of the diffracted beam is shaped by suitable beam limitation devices, before it is captured by a neutron detector. The gauge volume to which the strain measurement is related is given by the intersection of the incident and diffracted beams. Typically, neutron strain scanner is established on existing powder diffractometer. At the instrument of the first generation usually mosaic monochromator consisting of Cu, pyrolitic graphite or Ge is being used. As in the case of mosaic crystal monochromator there is no beam focusing, the uncollimated monochromatic beam spreads its cross-section at the place of the sample position up to tens of square centimetres. Then, the beam defining elements determining the gauge volume bring about a strong decrease of the final detector signal while the resolution (*FWHM* of the diffraction profile) is still strongly influenced by the mosaicity of the monochromator β being tens of minutes of arc. Even in the optimum case, when the diffraction angles at the monochromator (θ_M), and the sample (θ) fulfil the relation $\tan\theta / \tan\theta_M = +1/2$, *FWHM* has a minimum value but still it is larger than β [3]. As can be seen from Fig. 1, the best geometrical choice corresponds to 2θ=90° when the gauge volume is in the form of a cube or rectangular prism. Consequently, it is required to have an intense Bragg peak at $2\theta \approx 90°$. However, in the conventional case, due to the required resolution a large diffraction angle at the monochromator should be set. The current of the monochromatic neutrons impinging the sample is

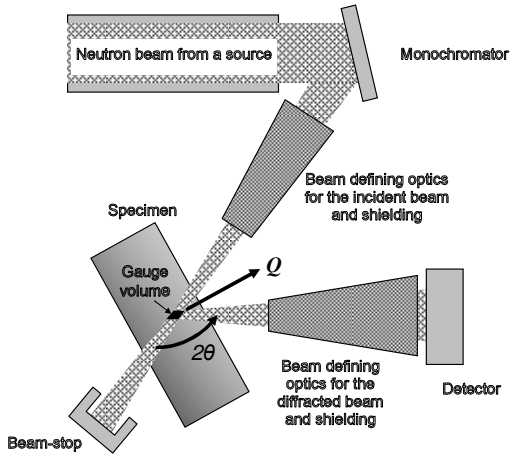

Fig. 1. Schematic illustration of a conventional reactor source based diffractometer for strain measurement.

proportional to the wavelength spread $\delta\lambda=\lambda\cdot\Delta\theta\cdot\cot\theta_M$, where $\Delta\theta$ is the angular divergence of the beam. Therefore, the larger diffraction angle at the monochromator means the smaller neutron beam current and consequently, the smaller detector signal. However, the situation is completely different in the case of employing focusing bent perfect crystal monochromator [4,5].

Focusing monochromator in the powder diffraction case

A great advantage of the strain diffractometer equipped with the focusing BPC-monochromator is very good predictability of its resolution and reflectivity parameters as has been many times demonstrated [6-10]. For imaging by a thin cylindrically bent crystal slab of the radius R, the lens formula

$$(f_a/a + f_b/b) = 1 \qquad (1)$$

can be used. The parameters $f_{b,a}=(R\cdot\sin(\theta_{hkl}\pm\Psi))/2$ are the focal lengths on the image or object side, respectively, which are dependent on the cutting angle Ψ (Ψ means the angle of the diffracting lattice planes with respect to the surface of the crystal slab). Symmetric geometry means $\Psi = 0$. The cutting angle Ψ can be also used as a free parameter in the instrument optimization procedure. In practice, the monochromator-sample distance L_{MS} is fixed and should coincide with the image

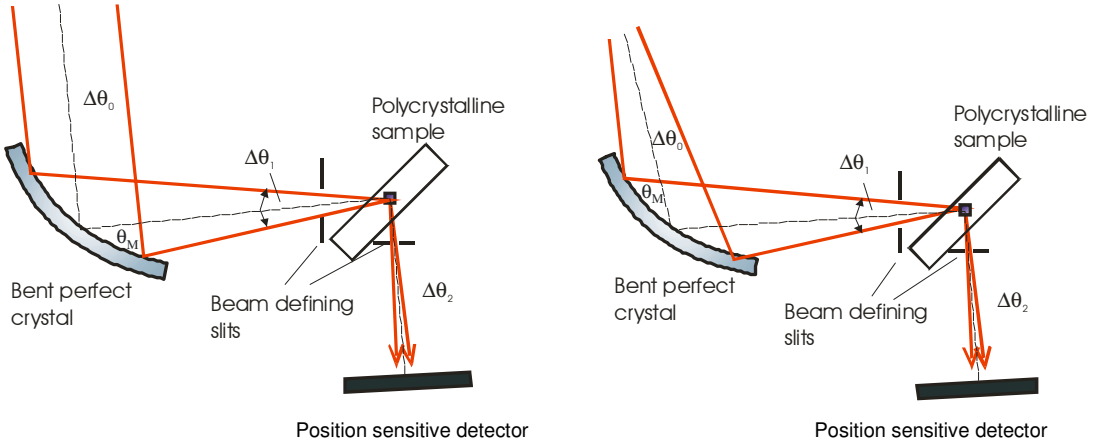

Fig. 2. Schematic geometry of the conventional diffractometer performance (left) and the one tested in KAERI (right)

distance b ($b = L_{MS}$) of a point source A being at a distance $a = L_{AM}$ before the monochromator. For the parallel incident beam is valid that $L_{MS} = f_b$ and $a = \infty$. Then, for given θ_M and the bending radius R for any point of the sample, we arrive at the relation

$$a(R_M) = f_b/(1 - f_b/b), \qquad (2)$$

where $f_b = (R/2)\sin\theta_M$ is the focal length. Notice, that each point of the sample is reached by a convergent bunch of strongly α–λ correlated rays according to

$$\Delta\lambda/\lambda = \Delta\alpha_1(1 - L_{MS}/2f_b)\cot\theta_M \ . \qquad (3)$$

Angular convergency (or divergency) $\Delta\alpha_0$ and $\Delta\alpha_1$ of the rays in the bunch before and behind the monochromator, respectively, are related as

$$\Delta\alpha_1 = 2\varepsilon(R) - \Delta\alpha_0, \qquad (4)$$

where $\varepsilon(R)=(W/R)\sin\theta_M$ is the total change of the angle of incidence (exit) over illuminated crystal length (W is the width of the incident beam in front of the crystal). Typical application of real space

focusing is that we focus the beam of a large cross-section (e.g. from a thermal neutron guide) on a small sample and thus transforming area into the solid angle.
The focusing diffraction performance (see Fig. 2) consists then of the following steps [8,9]: Monochromatic neutrons selected by the bent crystal from a white spectrum are focused on a sample (real space focusing). There is a strong correlation between the divergence of the incoming and outgoing beams with respect to the monochromator (see eq. (4)) and the sample as

$$\Delta\alpha_2=\Delta\alpha_1[2\ (\tan\theta_S / \tan\theta_M)(1 - L_{MS}/(R\sin(\theta_M-\Psi)) - 1]. \quad (5)$$

It can be seen from (5) that this correlation can easily be manipulated by changing the radius R. The indices S and M mean that the corresponding parameters are related to the sample and the monochromator, respectively. Then, by setting a radius of curvature of

$$R = (2L_{MS}/\sin(\theta_M-\Psi))/(2-1/a_{SM}), \quad (6)$$

the value of the divergence $\Delta\alpha_2$ equals to zero and in the vicinity of the chosen scattering angle $2\theta_S$ we obtain a (quasi)-parallel diffracted beam ($a_{SM} = -\tan\theta_S / \tan\theta_M$) as well as a highly luminous detector signal of the corresponding Bragg reflection. It also means that when the condition (6) is fulfilled, the resulted divergence is not dependent on $\Delta\alpha_1$ and a large width W of the polychromatic beam irradiated the monochromator can be successfully used. These are unique properties of the focusing diffractometer performance making a simultaneous increase of the detector signal and the diffraction profile resolution (decrease of its *FWHM*). It means in practice that depending on the crystal curvature, the peak intensity and *FWHM* of the diffraction line achieve their maximum and minimum, simultaneously. The quasi-parallel diffracted beam is then directly analyzed by using a position sensitive detector (PSD). It is clear from Fig. 2 that contrary to the conventional diffractometer performance with the mosaic monochromator no Soller collimators which always cut the neutron current are required.

Of course, there are small resolution uncertainties $\Delta\alpha_{2t}$ and $\Delta\alpha_{2w}$ influencing the instrumental resolution which make the (quasi-)parallel diffracted beam slightly divergent [9]. They come from a non-negligible thickness t_M of the monochromator and from the finite width w of the irradiated volume of the sample determined by the input and output slits. Thus, the monochromator of the thickness t_M and the width of the sample w bring about the uncertainties

$$\Delta\alpha_{2t} = 2t_M\, a_{SM} \cot\theta_M / R, \qquad \Delta\alpha_{2w} = w(2a_{SM} - 1)/L_{MS}\ . \quad (7)$$

Both of them bring about the diffracted beam slightly divergent (quasi-parallel) and directly determine the instrumental resolution. The uncertainty $\Delta\alpha_{2t}$ is in fact brought about by an effective mosaicity of the bent perfect crystal β' [11] which is usually several times smaller than the mosaicity of the conventional mosaic monochromator (also $\delta\theta << \varepsilon(R)$). Its value can be changed either by decreasing the crystal thickness t_M or similarly to the conventional performance by increasing the monochromator Bragg angle θ_M. However, it follows from (6) that the larger θ_M requires smaller R. On the other hand, $\Delta\lambda$ spread of the monochromatized beam decreases with the increasing the angle θ_M according to [3]

$$(\Delta\lambda/\lambda = \cot\theta_M \cdot \Delta\theta_1). \quad (8)$$

Therefore, the use of smaller monochromator Bragg angle θ_M, can contribute to an additional increase of the instrument luminosity. One more point should be taken into account that for $2\theta_M < 2\theta_S$, the divergence of the accepted beam impinging the monochromator $\Delta\alpha_0$ is not zero (see Fig. 2b) making a virtual source on the path between the reactor core and the focusing monochromator. The condition $\Delta\alpha_0 = 0$ is valid only for $2\theta_M = 2\theta_S$. Therefore, for $2\theta_M < 2\theta_S$ the use of any Soller collimator in the incident polychromatic beam is inadvisable. Even though that several

diffractometer parameters can be analytically calculated, it is clear that for full description of the diffractometer performance Monte Carlo simulations would be always desirable [12,13].

Experimental results

First, it was necessary to test the intensity and resolution behaviour of the diffractometer performance when using a small irradiated volume of a polycrystalline sample. For simplicity, instead of a bulk sample and beam defining slits we used an α-Fe(211) steel pin of 2 mm diameter and 40 mm height. By using a focusing Si(111) monochromator (cylindrically bent perfect crystal) set at $2\theta_M = 30^o$, for $\lambda = 0.162$ nm the scattering angle on the sample was $2\theta_S = 87.8^o$. In fact, several focusing diffraction geometries of the bent Si(111) monochromator were tested, both symmetric and asymmetric, and it was found that the symmetric one ist the best alternative (for details see our paper [14]). Fig. 3 shows the behaviour of the required characteristics of the

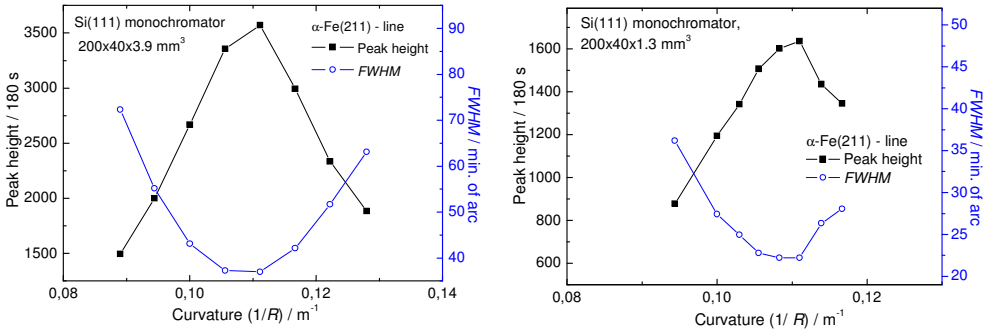

Fig. 3. The luminosity and resolution characteristics of the strain/stress diffractometer performance for the focusing Si(111) monochromator of different thickness of 3.9 mm (left) and 1.3 mm (right).

diffractometer on the monochromator crystal curvature. The resuls document the excellent property expected from the employment of the Bragg diffraction optics that the maximum luminosity is at the minimum of the resolution. Moreover, the resolution can be also optimized by a suitable choice of the thickness of the monochromator crystal slab (compare Fig. 3a and 3b) with an acceptable

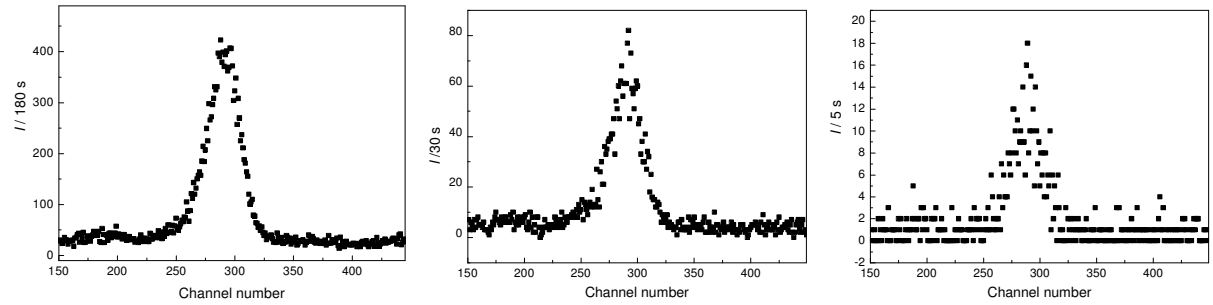

Fig. 4. Diffraction profiles of the sample of α-Fe(112) pin of 1 mm diameter and 40 mm height for different measurement times.

decrease of the detector signal. Fig. 4 displays the diffraction profiles of α-Fe(112) pin of 1 mm diameter and 40 mm height taken for different measurement times by the PSD at the distance of 120 cm from the sample. Smaller diameter of the samle has practically no influence on the *FWHM* of the diffraction proflile, because the spatial resolution of the detector was 2.5 mm (the angular width of one channel was 0.0095°) as well as the uncertainty $\Delta\alpha_{2t}$ were much larger. It is clear that by using a PSD having better spatial resolution, it could be set closer to the sample. In this case we used deliberately only 1 mm diameter pin as a sample and followed the value of the detector signal and the error in determination of the peak position. If we take into account that about 40 channels corresponds to the angular range corresponding to *FWHM,* even at the measurement time of 5 s the peak position can be determined with a relative error of about 10^{-4} which is sufficient in most cases

of residual strain/stress measurements. It follows from these facts that thanks to very good luminosity and resolution the new instrument performance permits also studies of some kinetic processes related to macro- and/or micro-strain/stress distribution. Namely, so called *in-situ* experiments with samples subjected to external termo-mechanical load can be attractive. In such cases much larger gauge volume is usually used which would permit to do the measurement time of the diffraction profile even shorter because the detector signal is roughly proportional to the irradiated volume of the sample,

Conclusion

The basic properties of unconventional strain/stress diffractometer performance employing Bragg diffraction optics are presented. It has been demonstrated that even at the medium power research reactor one can achieve very good luminosity and resolution of the dedicated instrument, which permit to carry out very effectively macro-strain/stress scanning but also micro-strain/stress studies and even to study some kinetic processes in polycrystalline materials running within a few seconds. Some improvements are still possible e.g. by installing horizontally and vertically focusing monochromator and a position sensitive detector od a better spatial resolution. As the vertical focusing has no influence on the resolution of the instrument, its future installation could improve the luminosity by a factor of 2-3.

Acknowledgement

This work was carried out under the Nuclear R&D Program by the Ministry of Education, Science and Technology of the Republic of Korea. Bragg diffraction optics investigations are in Czech Republic supported by research projects MSM2672244501, AVOZ104805505, ASCR-M100480901 and GACR P204/10/0654.

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