Development of the neutron diffraction method for stress measurements in thick steel samples.

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Abstract. The results of development of neutron method for stress measurements in bulk steel components are considered. Enhancement of the maximum available path length (~85mm) was achieved by using optimized bent perfect silicon crystal monochromators, position sensitive detectors and neutron wavelengths, corresponding to the minimums of neutron total cross section near Bragg edges.

1. Introduction

Thanks to high penetration ability of neutrons, the neutron diffraction method is widely used for nondestructive strain/stress measurements inside bulk polycrystalline materials [1]. The method is based on the accurate measurement of the diffraction peak position from a diffracting volume (gauge volume), defined by cadmium slits on the incident and diffracted beams. In a general case, three strain components along the “principal directions” should be measured to calculate stresses. Some components should be measured in “reflection geometry” and some components in “transmission geometry” (Fig 1).

Figure 1. (a) reflection geometry for the measurements of the normal (N) component; (b) transmission geometry for the measurements of the transverse (T) and longitudinal (L) components. The gauge volume is defined by slits in the incident (IS) and diffracted (DS) beams. The neutron path length (l) is the sum of the incident (lᵢ) and diffracted (lᵈ) beam path lengths.
Penetration ability of high energy synchrotron radiation is comparable with that for thermal neutrons, but there is difficulty in measuring strain components in “reflection geometry” because of a small (few degrees) scattering angle, \( \theta_S \) [2, 3]. Penetration ability of laboratory X-rays is much lower [2]. So, by now, only neutron method allows measuring all three strain components in bulk samples. However, because of low brightness of neutron sources, the maximum feasible path length in ferritic steel, \( l_m \), in modern neutron stress-diffractometers is limited to 60mm [3,4], that corresponds to 40mm thick plate. At the same time, development of heavy industries (e.g. shipbuilding, nuclear industry, chemical industry etc.) demands stress measurements in much thicker steel components.

This paper summarizes our results in development of neutron method for stress measurements in bulk steel components.

2. Experiment

The experiments were carried out at beam port ST-1 of HANARO reactor in KAERI, using the early and upgraded [5-8] versions of the residual stress instrument. The distance between the reactor core to the monochromator was 6.2 m; the monochromator–sample distance, \( L_{MS} \), was \( \sim 2.1 \) m; the sample to detector distance, \( L_{SD} \), was \( \sim 1.2 \) m. No Soller collimators were used. The open window of 40(\( w \))x70(\( h \))mm\(^2\) at 150mm from the drum center defined the beam impinging on the monochromator.

For testing the resolution and detector signal, the diffraction from the ferritic steel pin of diameter 2mm and height 40mm was measured. The pin was mounted in a sample position at the center of the diffractometer and imitated the gauge volume. For comparison of different experimental set-ups for stress measurements, we used the figure of merit, FoM, suggested in reference [9], which for the same acquisition time can be defined as

\[
\text{FoM} = \frac{I}{(\text{FWHM})^2 (\cot \theta_S)^2},
\]

where FWHM is the full width at half maximum of the diffraction peak in \( 2\theta_S \). The depth scan in reflection geometry of diffraction peaks 211, 110 of ferritic steel, and 311, 111 of austenitic steel, with gauge volume 80mm\(^3\) and measurement time 1h., was conducted to determine the maximal available path length, \( l_m \), defined as corresponding to the 100με (10\(^{-4}\)) strain error.

3. Results and discussion

3.1. Monochromator for a stress-diffractometer

In a stress-diffractometer at stationary reactor, it is necessary to scan only one diffraction peak at \( 2\theta_S \sim 90^0 \) [1]. A bent perfect silicon crystal (BPC) monochromator can be advantageous because its curvature can be adjusted to obtain maximal intensity and resolution of this particular peak [10-12]. Therefore, most of modern stress-diffractometers are equipped with double focusing bent perfect silicon crystal monochromators [13-16].

In experiments for monochromator optimization, we used a four-point type bending device, developed in NPI, Rez. [17], for cylindrical bending in horizontal plane of a perfect Si crystal slabs or “sandwiches” with reflecting area 40(\( w \))x200mm\(^2\) (\( w \)) and thickness up to 6 mm. We did not use vertical focusing because large divergence of the incident beam deteriorates definition of the vertical size of a gauge volume [1].

It was shown [18-22] that BPC monochromator at a take-off angle much smaller than 90\(^0\) can provide a much higher detector signal with excellent resolution in comparison to the mosaic crystal monochromators. Lattice planes (211), (110) of ferritic steel (b.c.c. structure) and planes (311), (111) of austenitic steel (f.c.c. structure) are recommended for strain measurements [1], because they are weakly affected by inter-granular strains. The wavelengths \( \lambda \sim 1.6\text{Å} \) for the reflections 211(b.c.c.) and 311(f.c.c.) and \( \lambda \sim 2.9\text{Å} \) for the reflections 110(b.c.c.) and 111(f.c.c.) are necessary for the diffraction peak at \( 2\theta_S \sim 90^0 \).
Our experiments with BPC monochromators [8,23-24] showed that for extracting monochromatic neutrons with wavelength $\lambda \sim 1.6\text{Å}$ the best choice (maximal FoM) is using BPC Si(220) monochromator with reflecting plane (220) in symmetric diffraction geometry at take-off angle $2\theta_M \approx 50^0$ (Table 1). Using Si(111) monochromator at smaller take-off angle $30^0$ significantly increases intensity, however, the broadening of peak results in lower FoM [7]. Using Si(113) monochromator at larger take-off angle $60^0$ increases resolution (decreases FWHM), however, loss in intensity results in lower FoM [24]. For comparison with Ge(311) vertical focusing mosaic crystal monochromator ($2\theta_M = 90^0, \lambda = 1.83\text{Å}$), the reflection 220 of austenitic steel pin (Ø2mm, h=40mm) was measured ($2\theta_0 = 90.4^0$) using Si(220) monochromator at take-off angle $2\theta_M = 57^0$ for the same wavelength.

Table 1. The figure of merit (FoM) for different monochromators, determined from measurements of diffraction peak 211 of ferritic and 220 of austenitic steel pins (Ø2mm, h= 40mm, t =180s.) [8, 23-24].

| Monochromator | Monochromator size | $2\theta_M^0$ | FoM |
|---------------|--------------------|---------------|-----|
| Si(111)-BPC   | 200x40x3.9 (mm³)   | 30            | 95  |
| Si(220)-BPC   | 200x40x3.9 (mm³)   | 50.6          | 100 |
| Si(113)-BPC   | 200x40x4 (mm³)     | 60            | 60  |
| Ge(311) mosaic, $\eta = 15^0$, vertical focusing | $60x220x8$ | 90 | 38 |

Neutrons with $\lambda \approx 2.9\text{Å}$ can be obtained using the Si(220) planes at $2\theta_M \approx 98^0$ or the Si(111) planes at $2\theta_M \approx 55^0$. The latter is preferable because at $2\theta_M \approx 55^0$ the instrument luminosity should be much higher than at $2\theta_M \approx 98^0$ [8], while a good resolution can be still achieved.

Thus, the Si(220) monochromator is preferable to extract “short” wavelengths near 1.6Å for the 211(b.c.c.) and 311(f.c.c.) reflections and the Si(111) monochromator to extract “long” wavelengths near 2.9 Å for 110(b.c.c.) and 111(f.c.c.) reflections.

It should be noted that in choosing the optimal resolution, both the FoM and the definition of gauge volume should be taken into consideration. Because of the divergence of the diffraction peak, the width of sampling gauge volume, $w$, is greater than the width of the cadmium slit on the diffracted beam, $s$. The difference depends on the slit to gauge volume distance, $l_{SG}$, and FWHM of a diffraction peak [25]. Estimation, made based on the results presented in [25], showed that FWHM= 0.4° is necessary for the 20% ($w/s = 1.2$) difference in widths for 2mm slit at $l_{SG} = 100$mm. Therefore, the diffraction peak width of FWHM=0.4° can be considered as reasonable for a stress diffractometer.

3.2. Position sensitive detector for a stress-diffractometer

At the scattering angle $2\theta_0 = 90^0$, the diffraction cone becomes a plane. Therefore, a position-sensitive detector (PSD), with a possibly high active area, increases the intensity without worsening the spatial and angular resolution [3]. However, a higher active area increases the uncertainty of the direction of the measured strain since the PSD subtends a larger vertical angle. We used a position sensitive detector, specially designed for RS measurements [26-27] with $200(h) \times 100(w) \text{mm}^2$ active area, 60% efficiency for $\lambda = 1.8$ Å, a delay line read-out mode, and spatial resolution of 2.5 mm. However, using 300mm high PSD with 2mm spatial resolution at distance $L_{SD} \sim 1$m [15-16] is preferable for stress measurements in thick samples, because it can additionally increase intensity, while uncertainty in strain direction $\pm 8^0$ is acceptable.
3.3. Wavelength selection
If a diffraction peak has a Gaussian shape and the background is negligible, the error in the strain determination, $Err(\epsilon)$, is [28-29]:

$$Err(\epsilon) = \frac{ucot\theta_S}{I^{1/2}},$$

where $u$ is the standard deviation of the peak profile in $\theta_S$ and $I$ is the integrated peak intensity. The integrated peak intensity ($I_l$) and the peak height ($H_l$) exponentially decrease with neutron path length, $l$, in material:

$$I_l = I_0 e^{-\sigma_t n_o l},$$

$$H_l = H_0 e^{-\sigma_t n_o l},$$

where $I_0$ and $H_0$ are the integrated peak intensity and the peak height for the zero path length, $\sigma_t$ is the total neutron-cross section per atom for single element material, $n_o$ is the number of atoms per unit volume and $l$ is the sum of the paths for the incident ($l_i$) and diffracted ($l_d$) beams ($l = l_i + l_d$, Fig. 1). At a certain depth the peak height becomes comparable with the background ($B_l$) and the error in strains ($Err(\epsilon)$) increases [30]:

$$Err(\epsilon)_l = \frac{\cot \theta_S u_\theta S}{(I_0 e^{-\sigma_t n_o l})^{1/2}} \left(1 + 2\sqrt{2} \frac{B_l}{H_0 e^{-\sigma_t n_o l}}\right)^{1/2}$$

The maximum available path length ($l_m$) can be increased by increasing gauge volume or measuring time. However, from (5) one can see that this is not effective because integral intensity exponentially decreases with neutron path length through the material [27]. A more effective way is to use neutrons with smaller total cross-section, $\sigma_t$, because it is in exponent.

In ferritic and austenitic steels, the main contribution to the neutron total cross section gives coherent Bragg scattering [31]. Therefore, the dependence of the total cross section on neutron wavelength has saw-tooth structure with abrupt changes at Bragg edges [32-34] (Fig. 2).

**Figure 2.** Calculated total neutron cross section of the ferritic steel (low-carbon steel, b.c.c.) and the austenitic steel (stainless steel 304L, f.c.c.) as a function of wavelength. Indices (hkl) of several Bragg edges and corresponding wavelengths are shown at the top. Scattering angles of reflections corresponding to the wavelength at Bragg edges are shown at the bottom. The wavelengths near minimums of cross sections marked with filled circles were tested.
Our experiments [34] showed that the maximum penetration path length in steels can be considerably increased (up to ~85mm) by using neutrons with wavelength $\lambda$, corresponding to the local minimum of neutron total cross section near but off the Bragg edges (Table 2).

**Table 2.** Maximum penetration path lengths ($l_m$) and depths in reflection ($D_{\text{ref}}$) and transmission ($D_{\text{tran}}$) geometries for different wavelengths in ferritic and austenitic steels [34] (2x2x20 = 80mm$^3$ gauge volume, 1h measurement time, $10^{-4}$ precision in strain).

| Monochromator | $\theta_M$(deg) | $\lambda$(Å) | Reflection plane | $\theta_S$(deg) | $l_m$(mm) | $D_{\text{ref}}$(mm) | $D_{\text{tran}}$(mm) |
|---------------|-----------------|--------------|-----------------|----------------|-----------|---------------------|---------------------|
| Si(220)       | 48              | 1.55         | (211)           | 82.9           | 77        | 26                  | 58                  |
| Si(111)       | 45              | 2.39         | (110)           | 72.1           | 83        | 24                  | 67                  |
| Si(220)       | 46.5            | 1.5          | (311)           | 87.8           | 75        | 26                  | 54                  |
| Si(220)       | 53.2            | 1.71         | (311)           | 104            | 76        | 30                  | 47                  |
| Si(111)       | 41.2            | 2.19         | (111)           | 63.8           | 87        | 23                  | 74                  |
| Si(111)       | 49.5            | 2.61         | (111)           | 78             | 89        | 26                  | 65                  |

The bent perfect crystal Si monochromator is advantageous for this purpose because the curvature can be optimized for the chosen wavelength in order to simultaneously achieve the maximum detector signal and the minimum peak width for a particular diffraction peak. The stress distribution in 50 mm thick welded ferritic steel plate [35] and dissimilar weld overlay pipe [36] were measured using this method (reflection 110 and wavelength $\lambda = 2.39\text{Å}$).

### 3.4. Two reflections/wavelengths method

Further development of this method [37-38] showed that available thickness of stress measurements in ferritic steel can be increased by using two different instrument configurations for measurement of different strain components. From Table 2, one can see that because of difference in scattering angles, the wavelength $\lambda = 2.39\text{Å}$ and the reflection 110 of ferritic steel ($\theta_S = 72.1^\circ$) are preferable for the measurement of the longitudinal (LD) and transverse (TD) strain components (transmission geometry), and the wavelength $\lambda = 1.55\text{Å}$ and the reflection 112 ($\theta_S = 82.9^\circ$) are preferable for the measurement of the normal (ND) strain components (reflection geometry).

In general, the calculation of stresses from strains measured with different hkl reflections is not correct because the diffraction elastic constants and inter-granular strains are hkl dependent. However, planes (211) and (110) of $\alpha$-Fe (bcc) have the same diffraction elastic constants calculated by the Kroner model: $E_{211,110} = 225.5\text{GPa}$, $\nu_{211,110} = 0.28$ [1]. Furthermore, planes (211) and (110) are less sensitive to the inter-granular strains and evident inter-granular effects in ferritic steel weld could not be observed [39]. The method was testified by measuring stress distribution in 50mm thick weld ferritic steel plate using one reflection 110 and two reflections 110, 211[37-38].

For stress measurements in 50 mm (and more), thick welded ferritic steel plates the following procedure was recommended: wavelength $\lambda = 2.39\text{Å}$ and reflection 110 for measurement of TD and LD components at all points through the thickness and ND components up to about 20mm depth. Wavelength $\lambda = 1.55\text{Å}$ and the reflection 112 were recommended for measurement of ND component in the remaining deeper locations. Using this “two reflections/wavelengths” method, the stress distribution in 70mm [40] and 80mm [4] thick welded ferritic steel plates were measured.
3.5. Magnetic field for stress measurement
As the neutron beam path increases in a ferritic steel specimen, significant peak broadening occurs, and this reduces the maximal available path length. Peak broadening was observed in ferritic steel and pure iron and was not observed in austenitic steel. Complementary neutron scattering experiments using small-angle neutron scattering (SANS) and wide-angle neutron diffraction (WAND) have been conducted [42] to investigate the reason of peak broadening. SANS experiments showed that applied magnetic field of 1.2T considerably decreases intensity of the small angle scattering in ferritic steel samples. It was supposed that peak broadening is caused by multiple refraction small angle scattering of neutrons on magnetic domain boundaries. The domain sizes calculated based on peak broadening (WAND) support the relationship between magnetic domains and peak broadening. Applied magnetic field of 0.5T increases maximum available path length over 80mm. Therefore it is supposed that the available neutron beam path length in ferritic steel can be considerably increased (~95mm) by applying sufficiently strong (~1.5T) magnetic field.

4. Conclusion
In the last 10 years, the neutron method achieved significant progress in increasing maximal available depth for strain measurements. It was possible thanks to development of dedicated bent perfect crystal silicon monochromators and position sensitive detectors. Further enhancement of available depth in steels was achieved by using neutrons with wavelengths, corresponding to the minimums of the neutron total cross section near but off Bragg edges. Measurement of stress distribution in 80mm thick steel plate is now possible. The maximum available neutron beam path length in ferritic steel can be additionally increased by applying sufficiently strong magnetic field.

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