The neutron texture diffractometer at the China Advanced Research Reactor*

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Abstract: The first neutron texture diffractometer in China has been built at the China Advanced Research Reactor, due to strong demand for texture measurement with neutrons from the domestic user community. This neutron texture diffractometer has high neutron intensity, moderate resolution and is mainly applied to study texture in commonly used industrial materials and engineering components. In this paper, the design and characteristics of this instrument are described. The results for calibration with neutrons and quantitative texture analysis of zirconium alloy plate are presented. The comparison of texture measurements with the results obtained in HIPPO at LANSCE and Kowari at ANSTO illustrates the reliability of the texture diffractometer.

Keywords: neutron diffraction, neutron texture diffractometer, bulk texture, industrial application

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1 Introduction

Usually, the orientations of crystallites in polycrystalline materials have some preferred direction. The structure with this preferred orientation is known as the texture of the polycrystalline material [1]. As an intrinsic feature of metals, ceramics and polymers [2], texture strongly affects many properties of polycrystalline materials [3], and is seen as one of the most important features to fully characterize any kind of polycrystalline material at the microstructural level. Usually, the texture distribution can be quantitatively described by the orientation distribution function (ODF) with respect to a macroscopic sample coordinate system.

Texture determination is usually based on the pole figure measurements by X-ray, synchrotron radiation, electron back-scatter diffraction and neutron diffraction. Among these techniques, neutron diffraction has its own advantages due to the low absorption coefficient of neutrons [3, 4] (the penetration depth for most materials in neutron diffraction is a factor of 10^2-10^3 larger than for X-ray diffraction). These advantages include:

1) Large samples can be used, so high accuracy and good statistics as well as bulk texture can be obtained;

2) Accurate determinations of texture can be achieved for coarse-grained, inhomogeneous textured, multiphase samples and for samples with a small volume fraction of second phase;

3) The scattering length of neutrons changes irregularly with atomic number, even for various isotopes of the same element, so the texture studies of light element phases, especially in the presence of other phases containing heavy elements, is much easier;

4) Neutrons can be scattered magnetically - this effect is used for magnetic texture analyses;

5) More importantly, texture measurements for samples in non- ambient environments such as high temperature and load can easily be carried out.

Among the texture analysis methods, "Texture analysis by neutron diffraction has become a standard method to investigate bulk textures of various materials" [5]. In consequence, more than ten neutron texture diffractometers have been built in different neutron scattering laboratories around the world.

However, like other neutron scattering experiments, pole figure measurement by neutrons requires an intense

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neutron source such as a nuclear reactor with at least medium neutron flux, or a spallation neutron source. The China Advanced Research Reactor (CARR) at the China Institute of Atomic Energy (CIAE), with a maximum thermal neutron flux of about 8×10^{14}/sec·cm^2[6, 7], is well qualified for neutron texture measurements and other neutron scattering experiments. A number of different types of neutron scattering instruments have now been or are being built around the reactor. The Neutron Texture Diffractometer (NTD) is one of these instruments and the first platform for neutron texture measurement in China. In order to make full use of this instrument by the international and domestic user community, the design, characteristics and performance of the newly constructed NTD are introduced in this paper.

2 Instrument design and characteristics

The NTD is located at beam tube H2-1 of CARR. Its main components and layout are shown in Fig. 1.

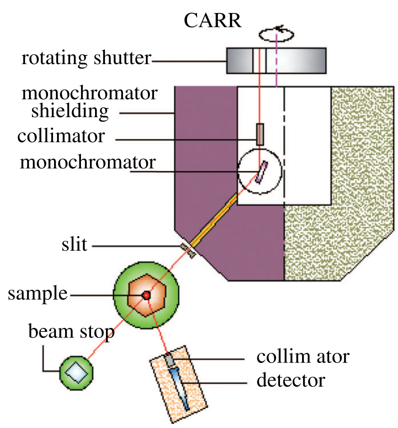

Fig. 1. (color online) Layout of neutron texture diffractometer at CARR.

The design was developed based on the considerations of neutron intensity and instrument resolution. Since pole figure measurements are very time-consuming, the most important factor for a texture diffractometer is to have high neutron intensity. On the other hand, high resolution means that more well-separated reflection peaks can be chosen and samples with larger cells and more complicated structures can be investigated [8, 9].

According to Caglioti, the instrument resolution function as expressed by $A_{1/2}$, the full width at half maximum for the peaks, and the integrated peak intensity L of the instrument are given as follows [10–12]:

$$A_{1/2}^2 = U\tan^2\theta + V\tan\theta + W,$$

$$U = \frac{4(\alpha_1^2\alpha_2^2+\alpha_1^2\beta^2+\alpha_2^2\beta^2)}{\tan^2\theta_{\rm M}(\alpha_1^2+\alpha_2^2+4\beta^2)},$$

$$V = \frac{-4\alpha_2^2(\alpha_1^2+2\beta^2)}{\tan\theta_{\rm M}(\alpha_1^2+\alpha_2^2+4\beta^2)},$$

$$W = \frac{\alpha_1^2\alpha_2^2+\alpha_1^2\alpha_3^2+\alpha_2^2\alpha_3^2+4\beta^2(\alpha_2^2+\alpha_3^2)}{\alpha_1^2+\alpha_2^2+4\beta^2}, \quad (1)$$

$$L = \frac{\alpha_1\alpha_2\alpha_3\beta}{(\alpha_1^2+\alpha_2^2+4\beta^2)^{1/2}}, \quad (2)$$

where α_1, α_2 and α_3 are the horizontal divergences of the primary, second and third collimators, respectively, β is the monochromator mosaic spread, and $2\theta_{\rm M}$ is the take-off angle of the monochromator.

As can be seen, the above two requirements are in conflict with each other. The instrument resolution can be easily improved by using collimators with smaller horizontal divergence, but this will drastically decrease the diffractometer intensity. It is not possible for a texture diffractometer to achieve both resolution and intensity as high as desired.

If the instrument resolution function is designed higher than that required for the samples to be measured, a meaningless loss in diffraction intensity will be caused, which is unacceptable due to the fact that the reactor sources available in the world are not strong enough for neutron scattering experiments. Therefore a compromise has to be made. In the case of NTD at CARR, most of the materials to be measured have simple structures and relatively small cells. For the pole figure measurements of these materials, the instrument is required to have only moderate resolution, which enables it to achieve high neutron intensity. Obviously, in order to make the instrument reasonable and efficient, the best solution is to let the instrument resolution function just match the required one in a large enough 2θ range.

To obtain the required resolution function, the expression for the separation between adjacent peaks for cubic cells deduced from the Bragg equation was used appropriately in the design (see Eq. (3)). Here, λ is the neutron wavelength, 2θ is the scattering angle and $a$is the size of the unit cell. It is clear that the required resolution function is symmetrical about the minimum $2\theta=90°$.

$$\Delta(2\theta) = 2\Delta\theta = \left(\frac{\lambda}{2a}\right)^2\frac{2}{\sin 2\theta}. \quad (3)$$

In practice, the monochromator was selected before the "matching" procedure so that the parameters λ, $2\theta_{\rm M}$ and β for the instrument were determined first. A single crystal used as monochromator should have high neutron reflectivity, appropriate mosaic spread β for high neutron intensity and a proper plane spacing d for the take-off angle $2\theta_{\rm M}$ and the reflected neutron wavelength λ required by a specific spectrometer.

Unfortunately, one has only a few choices since the growth of single crystals large enough for use is very difficult. For our texture diffractometer, a Cu(111) single crystal from the previous Four Circle Diffractometer

(FCD) at the Juelich Center for Neutron Science (JCNS) is used as the monochromator. Single crystals of Cu have relatively high neutron reflectivity and are often used as monochromators in neutron scattering instruments, but the mosaic spread β of this Cu(111) crystal is only about $6'$, which is too small for a texture instrument from the point of view of intensity, although the lower β value benefits the resolution for angles below the minimum $A_{1/2}$ position. Fortunately, it is a vertically bent monochromator, so the intensity gain factor of 2–3 can compensate the intensity loss due to the too small β. This instrument will be equipped with a double focusing HOPG(002) or Si(311) monochromator in the future.

With the known interplanar spacing of Cu(111), d=2.0871Å, the neutron wavelength λ can be obtained from the Bragg equation $2d\sin\theta_{\mathrm{M}} = \lambda$. Usually, the take-off angle $2\theta_{\mathrm{M}}$ is set to about 2θ=90°, or even a larger angle, so that a good match between the instrument and required resolution functions can be obtained over a larger 2θ angle range. However, for our instrument the maximum $2\theta_{\mathrm{M}}$ we can choose is restricted to about $2\theta_{\mathrm{M}}$=40° due to the limited geometric space, and thus the corresponding neutron wavelength becomes $\lambda \approx$1.43Å. Although the use of this low take-off angle will lower the resolution of the instrument, it is still acceptable for most texture measurements, whereas the corresponding neutron wave length λ is near the maximum of the incident neutron wave length spectrum, and thus is beneficial to the intensity [13, 14].

With the given λ, β and $2\theta_{\mathrm{M}}$, the range of the required resolution functions was estimated through calculating the required resolution functions for several typical metals and then a series of calculations were carried out for the instrument resolution function for different combinations of α_1, α_2 and α_3.

Usually, in order to obtain high neutron intensity with relatively small sacrifice of resolution, a large angle α_2 is chosen. In our case, the second collimator is needless since the natural collimation caused by the geometric layout of the instrument of $\sim 30'$ can fully replace the use of α_2. Therefore, all the calculations for the instrument resolution function were performed with α_2 equal to $30'$. By careful comparisons between the calculated instrument and required resolution functions, three different combinations of $\alpha_{1,}\alpha_3$ were finally adopted to meet the requirements of different materials. The results are listed in Table 1 and have been used in the construction of the texture diffractometer. The corresponding instrument resolution functions are plotted in Fig. 2.

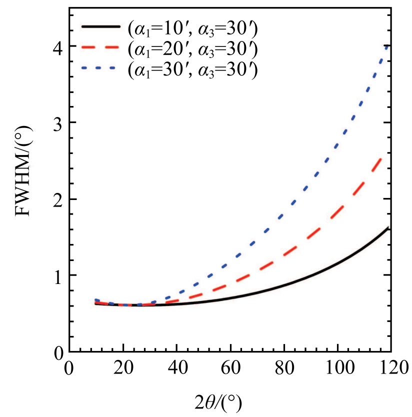

Fig. 2. (color online) Instrument resolution functions for different combinations of α_1 and α_3.

Besides the monchromator and collimators, the sample table is another important component of the texture diffractometer, although it does not affect the intensity and resolution directly. For our instrument, the four-circle mechanical device of the above-mentioned Juelich FCD was modified and reused as the sample table. With a four-circle device, the pole figure $P_{\mathrm{hkl}}(\alpha, \beta)$ for a specified plane (hkl) can be obtained by rotation of the sample through the χ (0°−90°) and φ(0°−360°) axes with a certain step size to align the various sample orientations along the (hkl) scattering vector, which is usually the bisector of the incident and the diffracted beam, and to record the diffracted intensities at each step using a detector set at the 2θ position of the (hkl) reflection.

For the pole figure measurement to be realized, the hardware and software for motion control and data acquisition were specially designed by Xiaolong Liu et al. [15]. Shown in Fig. 3 is the hardware structure. The software was written in the Python language under the Linux system, and has been proved to be reliable by practical pole figure measurements.

It is worth mentioning that the ^{3}He single detector used at present will be replaced in the near future by a 200 mm×200 mm two-dimensional position sensitive detector (PSD) funded by the International Atomic Energy Agency. With this PSD, it is possible to measure several pole figures simultaneously. Moreover, overlapping pole figures may be separated, which is especially important for materials with line-rich diffraction patterns, such as

Table 1. Characteristics of NTD at CARR.

maximum beam size	collimator	monochromator	take-off angle	wavelength	neutron flux at sample	detector
30 mm×30 mm	$\alpha_1 = 10', 20', 30'$; $\alpha_3 = 30'$	vertical bent Cu(111)	42°	1.48 Å	5.6×10^7 n cm^{-2}s^{-1}	PSD; ^{3}He tube

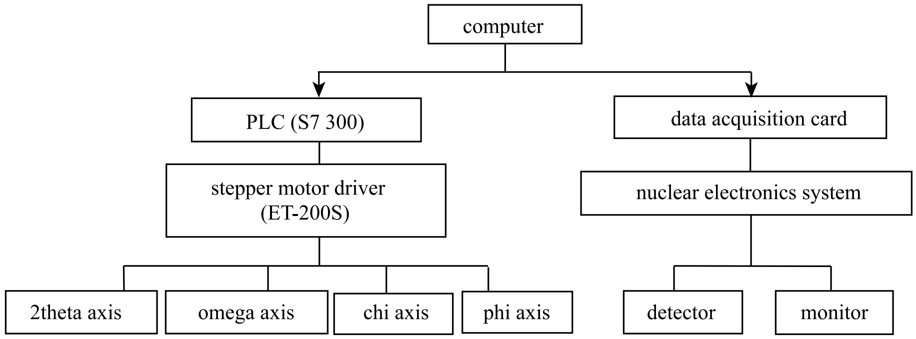

Fig. 3. Hardware structure of motion control and data acquisition.

intermetallic phases, ceramics and composites. The characteristics of this instrument are given in Table 1.

3 Experimental measurements

3.1 Diffraction pattern measurement for sample TiO_2

The diffraction pattern of a cylindrical TiO_2 sample with height 20 mm and diameter 10 mm was measured with $\alpha_1 = 30'$ to calibrate this instrument. The sample was mounted on a thin Al rod wrapped by a Cd foil and inserted into the center of the goniometer. The neutron beam size is 20 mm (width) $\times$ 30 mm (height). A 2θ-scan in the range of 25° to 72° was carried out at a step of 0.1°. The Rietveld profile technique was used to analyse the data with the program Fullprof, using the known structure of TiO_2.

Figure 4 shows the measured and calculated diffraction patterns of the TiO_2 sample. As can be seen, a good fit between them was achieved. The actual neutron wavelength λ and the corresponding take-off angle $2\theta_M$ thus obtained, are listed in Table 2. The real resolution curve of the instrument derived from the fit is plotted in Fig. 5, together with the designed resolution curve. Good agreement between them is also found.

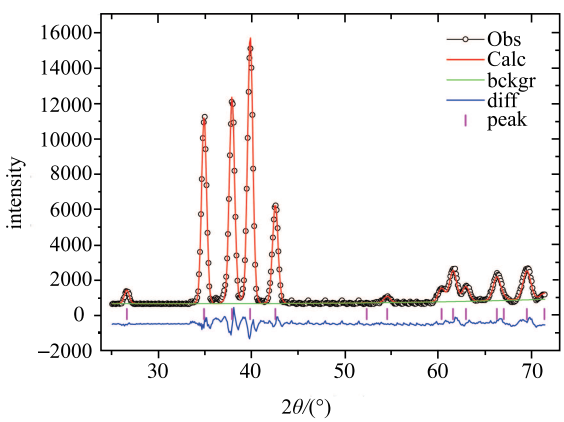

Fig. 4. (color online) Neutron diffraction pattern of TiO_2 sample.

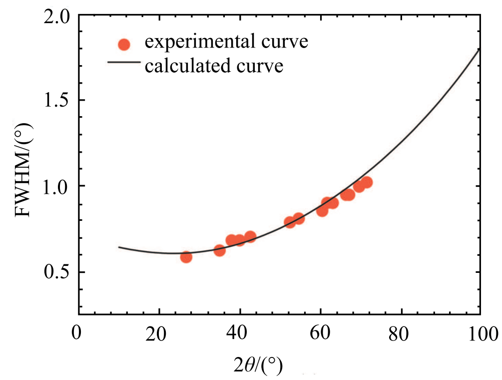

Fig. 5. (color online) Resolution curve of the texture diffractometer at CARR.

3.2 Neutron intensity measurement

The gold foil activation method was adopted to measure the neutron flux at the sample position. A circular gold foil with diameter 19 mm was fixed at the center of the sample table, and totally bathed in the neutron beam for about 6 h at the reactor power of 10 MW. Through measuring the actively of ^{198}Au produced by the reaction of ^{197}Au(n,γ) ^{198}Au, the neutron flux was obtained. Corresponding to a reactor power of 60 MW, it is 5.6×10^7 n cm^{-2}s^{-1} at the sample position, and is also listed in Table1.

3.3 Pole figure measurements for warm-rolled Zircaloy-4 plate

The pole figures of a round-robin sample of warm-rolled Zircaloy-4 were measured to test the performance of this texture diffractometer. The texture measurements for this sample have also been carried out at the neutron texture diffractometers of High-Pressure-Preferred Orientation (HIPPO) at LANSCE and Kowari at ANSTO. A cubic sample of 12 mm$\times$12 mm$\times$12 mm was prepared by spark cutting a warm-rolled Zircaloy-4 plate into small squares, and then gluing them together along the same rolling direction. The size of incident neutron beam was set as 25 mm$\times$25 mm by the variable slit to ensure the sample bathing in the neutron beam.

Table 2. Kearns factors and texture index for the Zircaloy-4 specimens.

instrument	transverse	normal	rolling	sum	text index
CARR-TD	0.352	0.527	0.118	0.997	2.02
HIPPO	0.359	0.543	0.096	0.998	2.15
kowari	0.357	0.551	0.090	0.998	2.32

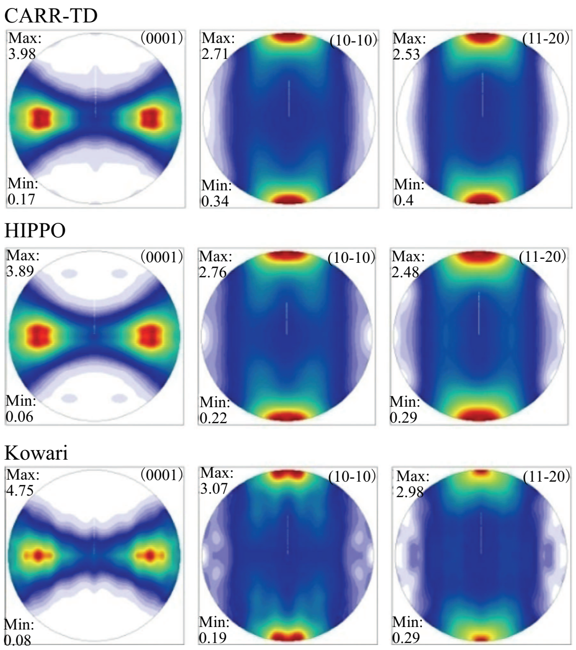

Fig. 6. (color online)(0001), (10-10) and (11-20) pole figures recalculated from the ODF of warm-rolled Zircaloy-4 sample.

The (10-10), (0002), (10-11) and (11-20) pole figures were each measured at the reactor power of 10 MW. The time taken for each pole figure was about 4 h. The raw data were transferred into LaboTex format, then used for quantitative ODF analysis. Shown in Fig. 6 are the (0001), (10-10) and (11-20) pole figures, recalculated by J.R. Santisteban from the obtained ODF together with those measured at HIPPO and Kowari. Good agreement is found both qualitatively and quantitatively between the different instruments [16]. The Kearns factors and texture indices were also calculated, and the results are shown in Table 2. All Kearns factors obtained on different instruments are within an uncertainty of ± 0.02, which is lower than typical differences usually found for different batches, or between the start and end of pressure tubes [17].

4 Summary

The newly constructed neutron diffractometer at CARR is the first instrument in China for texture pole figure measurement by neutrons. With relatively high neutron intensity and performance, it has been proved to be efficient and suitable for pole figure measurements for simple structure materials in the present stage. Up to now, a number of pole figures with satisfying quality have been obtained on this instrument for different research projects. A further improvement in its performance will be made in the near future by replacing the single ^{3}He detector with a two dimensional PSD.

The NTD at CARR has been opened to the user community. Users are welcome to apply to use this instrument for their research projects independently or in cooperation with us.

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