The 7th International Topical Meeting on Neutron Radiography

Upgrade of Bragg edge analysis techniques of the RITS code for crystalline structural information imaging

H. Sato\textsuperscript{a,b,*}, T. Shinohara\textsuperscript{a}, R. Kiyana\textsuperscript{a}g, K. Aizawa\textsuperscript{a}, M. Ooi\textsuperscript{a}, M. Harada\textsuperscript{a}, K. Oikawa\textsuperscript{a}, F. Maekawa\textsuperscript{a}, K. Iwase\textsuperscript{c}, T. Kamiyama\textsuperscript{b} and Y. Kiyana\textsuperscript{b}

\textsuperscript{a}J-PARC Center, Japan Atomic Energy Agency (JAEA), 2-4 Shirakata-shirane, Tokai, Naka, Ibaraki 319-1195, Japan
\textsuperscript{b}Faculty of Engineering, Hokkaido University, Kita-13 Nishi-8, Kita-ku, Sapporo, Hokkaido 060-8628, Japan
\textsuperscript{c}Frontier Research Center for Applied Atomic Sciences, Ibaraki University, 162-1 Shirakata, Tokai, Naka, Ibaraki 319-1106, Japan

Abstract

The RITS code was developed for quantitative evaluation and visualization of crystalline structural information (e.g. crystallographic texture, preferred crystal orientation, crystallite block size and crystal lattice strain, etc.) of a material, analyzing position-dependent Bragg edge transmission spectra measured in an imaging experiment with pulsed neutrons. Originally, this code was neither able to deal with all the crystal structures that were classified into 230 types of space groups, nor applicable to materials composed of multi elements and multi crystalline phases. Therefore, we improved the RITS code for expansion of its capability, and then performed some demonstrations of simulation calculations and profile fitting analyses of experimental data. In this article, we present the new algorithm and the new functions of the improved RITS code, and the results of the simulation calculations and the experimental data analyses.

© 2013 The Authors. Published by Elsevier B.V. Open access under CC BY-NC-ND license.
Selection and/or peer-review under responsibility of ITMNR-7

Keywords: pulsed neutron imaging; Bragg edge; RITS; Crystal structure; Crystalline phase

* Corresponding author. Tel.: +81-11-706-6679; fax: +81-11-706-6679.
E-mail address: h.sato@eng.hokudai.ac.jp.

1875-3892 © 2013 The Authors. Published by Elsevier B.V. Open access under CC BY-NC-ND license.
Selection and/or peer-review under responsibility of ITMNR-7
doi:10.1016/j.phpro.2013.03.022
1. Introduction

The RITS (Rietveld Imaging of Transmission Spectra) code [1-3] was developed for the spectral analyses of wavelength- and position-dependent “Bragg edge” transmission data obtained in imaging experiments by the time-of-flight (TOF) method of pulsed neutrons. Since the RITS code can quantitatively evaluate crystalline structural information at each position of a specimen, the quantitative visualization of such information of a structural or functional material can be achieved. Fig. 1 shows an example of the Bragg edge transmission pattern caused by diffraction phenomenon of neutrons in an alpha-iron plate of 5 mm thickness. Bragg edges include various crystallographic information; the whole pattern (edges’ positions and heights) reflects the crystal structure and the crystalline phase, the shape change of the sawtooth of Bragg edge transmission indicates the crystallographic texture and the preferred crystal orientation [1-3], the intensity increase caused by multiple scattering of neutrons inside the same crystallographic mosaic block includes information of the crystallite block size [2,3], and the wavelength position of a Bragg edge corresponds to two times of the crystal lattice plane spacing including the crystal lattice strain information [4,5]. Such information is important for controlling the mechanical properties of engineering materials, and the imaging technique can gain the information by non-destructive way with a high spatial resolution over the wide area of a bulk specimen. Therefore, the pulsed neutron imaging using the Bragg edge transmission spectroscopy is expected as one of the most useful analysis tools for materials research besides the electron microscope, the X-ray/neutron scattering techniques and the synchrotron phase-contrast microtomography.

The RITS code is indispensable for quantitative evaluation of crystallographic information included in pulsed neutron transmission data. However, the RITS code was not applicable to all the crystal structures that were classified into 230 types of space groups. Additionally, the RITS code also was not able to deal with the transmission data of materials composed of multi elements and multi crystalline phases. The capability of the RITS code was not satisfactory for material researchers. Therefore, in this study, we improved the functions of the RITS code in order to solve these problems, and then demonstrated the simulation calculations and the spectral profile fitting analyses for the verification.

![Fig. 1. Bragg edge transmission spectrum as a function of neutron wavelength, and crystalline structural information included in the spectrum. Since the spectrum like this can be measured at each pixel of a neutron imaging detector, we can achieve the quantitative crystallographic information visualization of a material owing to using the spectral analysis code, RITS.](image-url)
2. Algorithm of the improved RITS code

In this section, we present new algorithm of the improved RITS code that can be applicable to data analysis of the pulsed neutron Bragg edge transmission spectrum of a material composed of every crystal structure, multi elements and multi crystalline phases.

The neutron transmission spectrum as a function of neutron wavelength \( \lambda \), \( Tr(\lambda) \), is represented by the Beer-Lambert-Bouguer law, as follows:

\[
Tr(\lambda) = \exp \left( - \sum_p \sigma_{\text{tot},p}(\lambda) \rho_p t_p \right),
\]

where \( \sigma_{\text{tot},p}(\lambda) \) is the microscopic neutron-nucleus total cross section, \( \rho_p \) is the atomic number density, and \( t_p \) is the effective transmission thickness of each crystalline phase \( p \). In this article, we deal with \( \rho^\times t \) as the projection data of the atomic number density or the projected atomic number density. The value of the projected density \( \rho^\times t \) of each phase \( p \) is the refinable parameter during the profile fitting between experimental data and simulation data, quantitatively evaluated for the crystalline phase imaging. The improved RITS code can deal with not only the condition of single crystalline phase \( p = 1 \) but also the condition of multi crystalline phases \( p > 1 \).

The total cross section \( \sigma_{\text{tot}}(\lambda) \) consists of elastic coherent scattering, elastic incoherent scattering [6], inelastic coherent scattering [6-9], inelastic incoherent scattering [6-9] and absorption (following the \( 1/\nu \) law) components, as follows:

\[
\sigma_{\text{tot}}(\lambda) = \sigma^{\text{ela}}_{\text{coh}}(\lambda) + \sigma^{\text{ela}}_{\text{incoh}}(\lambda) + \sigma^{\text{inela}}_{\text{coh}}(\lambda) + \sigma^{\text{inela}}_{\text{incoh}}(\lambda) + \sigma_{\text{abs}}(\lambda).
\]

In the RITS code, the elastic coherent scattering cross section is calculated by the kinematical diffraction theory [10] combined with the Jorgensen type edge profile function \( R_{hkl}(\lambda-2d_{hkl}) \) [11], the March-Dollase preferred orientation function \( P_{hkl}(\lambda, 2d_{hkl}) \) [12] and the Sabine’s primary extinction function \( E_{hkl}(\lambda, 2d_{hkl}, F_{hkl}) \) [13], as follows:

\[
\sigma^{\text{ela}}_{\text{coh}}(\lambda) = \frac{\lambda^2}{2V_0} \sum_{hkl} \left| F_{hkl} \right|^2 d_{hkl} R_{hkl}(\lambda-2d_{hkl}) P_{hkl}(\lambda, 2d_{hkl}) E_{hkl}(\lambda, 2d_{hkl}, F_{hkl}),
\]

where \( V_0 \) is the unit cell volume of the crystal lattice, \( F_{hkl} \) is the crystal structure factor including the Debye-Waller factor, and \( d_{hkl} \) is the crystal lattice plane spacing. The crystal structure factor \( F_{hkl} \) is calculated by

\[
F_{hkl} = w_{hkl} \sum_n o_n b_n \exp \left[ 2\pi i (hx_n + ky_n + lz_n) \right] \exp \left( -\frac{B^\text{iso,n}}{4d^2_{hkl}} \right).
\]

Here, \( w_{hkl} \) is the multiplicity, \( o_n \) is the site occupancy, \( b_n \) is the coherent scattering length, \( (x_n, y_n, z_n) \) is the fractional coordinates, and \( B^\text{iso,n} \) in the Debye-Waller factor is the isotropic atomic displacement parameter of \( n \) th site atom in the crystal lattice, respectively.

In the improved RITS code, a new subroutine was implemented for automatic calculation of the crystal structure factor of all the crystal structures. This calculation component reads the space symmetry control parameters (the rotation matrices \( R \) and the translation vector \( T \) ) from the database containing the information of the coordinates of equivalent positions of 230 space groups, and can automatically give the all atomic coordinates in the crystal lattice by using only the \( n \) th site fractional coordinates \( (x_n, y_n, z_n) \) information with the equivalent coordinates information. Then, \( s \) th equivalent position of \( n \) th site general position is given by
\[
\begin{pmatrix}
    x_{ns} \\
    y_{ns} \\
    z_{ns}
\end{pmatrix}
= \begin{pmatrix}
    R_{11s} & R_{12s} & R_{13s} \\
    R_{21s} & R_{22s} & R_{23s} \\
    R_{31s} & R_{32s} & R_{33s}
\end{pmatrix}
\begin{pmatrix}
    x_n \\
    y_n \\
    z_n
\end{pmatrix}
+ \begin{pmatrix}
    T_{1s} \\
    T_{2s} \\
    T_{3s}
\end{pmatrix}
= \begin{pmatrix}
    R_{11s}x_n + R_{12s}y_n + R_{13s}z_n + T_{1s} \\
    R_{21s}x_n + R_{22s}y_n + R_{23s}z_n + T_{2s} \\
    R_{31s}x_n + R_{32s}y_n + R_{33s}z_n + T_{3s}
\end{pmatrix}
\tag{5}
\]

from the user-manual of the Rietveld analysis code for powder diffractometry, RIETAN-FP [14]. Owing to this new calculation component, the RITS code can automatically calculate the crystal structure factor of 230 types of space groups. At the same time, this technique also can deal with a crystal structure composed of multi elements. Thus, the RITS code can be applicable to the transmission data of all the crystal structures, multi elements and multi crystalline phases.

3. Verification results of the new calculation algorithm for the crystal structure factor

In this section, we present some simulation calculation results of the neutron-nucleus total cross section obtained by the improved RITS code.

Fig. 2 shows simulation calculation results of microscopic total cross section of (a) alpha-iron having the body-centered-cubic (BCC) crystal structure classified as the space group number 229, (b) nickel having the face-centered-cubic (FCC) crystal structure classified as the space group number 225, (c) magnesium having the hexagonal-close-packed (HCP) crystal structure classified as the space group number 194, and (d) multi elemental hydrogen storage alloy Ti\textsubscript{0.45}Cr\textsubscript{0.25}Mo\textsubscript{0.30} having the BCC crystal structure. Firstly, the data of only Fig. 2 (a) and (b) have been calculable for the previous RITS code.
Both calculation results of the improved and previous RITS codes correspond to each other for Fig. 2 (a) and (b). Furthermore, Fig. 2 (c) correctly corresponds to the typical powder diffraction pattern of the HCP crystal structure although this type of the crystal structure was not able to be automatically calculated by the previous RITS code.

\( \text{Ti}_{0.45} \text{Cr}_{0.25} \text{Mo}_{0.30} \) is known as a null alloy for coherent scattering of neutrons. Actually, Fig. 2 (d) correctly indicates that the Bragg diffraction (the elastic coherent scattering) is tiny due to the negative scattering length of Ti. Additionally, it is successfully predicted that the inelastic scattering is emphasized at the high energy region. Thus, it is verified that the improved RITS code implementing the Eq. (5) algorithm can successfully operate as well as the previous RITS code.

4. Quantitative evaluation and imaging of a multi crystalline phases material

For quantitative crystalline phase imaging, it is necessary to quantitatively evaluate the quantity of each phase. In the improved RITS code, the projected atomic number density (atomic number density ×...
effective transmission thickness) $p^\times t_p$ of each phase can be evaluated as described in Sec. 2. Then, we demonstrated the crystalline phase imaging by analyzing experimental position-dependent Bragg edge transmission data of a material composed of two crystalline phases.

4.1. Experimental

The detailed information of the experiment has been already described in Ref. [15]. The experimental data were measured at BL20 (the beam-line for the versatile powder diffractometer “iMATERIA”) at Materials and Life Science Experimental Facility (MLF) at Japan Proton Accelerator Research Complex (J-PARC) in Japan. The neutron imaging detector was the $^6$Li-glass scintillator pixel-type detector ($8\times8$ grid type) [16,17]. The pixel size is 2.3 mm×2.3 mm, and the detection area is 18.4 mm×18.4 mm. The detection efficiency is almost 100 % for cold neutrons because the scintillator thickness is 1 mm. The exposure time was 3 hours. This is because the J-PARC accelerator was unstable, and the proton accelerator beam power was only 4 kW during this experiment.

Fig. 3 shows a photograph of the measured specimen. The sample was a welded plate composed of JIS-SS400 (alpha-iron, the BCC crystal structure) and JIS-SUS304 (austenite stainless steel Fe-9%Ni-19%Cr, the FCC crystal structure) of 6 mm thickness. This plate has the welded zone along the center line, and also the heat affected zone (HAZ) around the welded zone.

4.2. Results and discussion

Fig. 4 shows examples of the profile fitting analysis of experimental one pixel data at (a) only alpha-iron (the BCC phase) zone, (b) only austenite stainless steel (the FCC phase) zone, and (c) both the BCC and FCC phases zone. The statistical errors were about 1 % at the energy region of cold neutrons. These experimental data with the fitting curves indicate that the profile fitting of not only the single phase data but also the two phases data can be successfully carried out by the improved RITS code. Owing to such spectral analysis, we can refine the crystalline structural parameters in the RITS code for the experimental data, and then can quantitatively evaluate the crystalline structural information at each zone.

Fig. 5 (a-1) and (b-1) show the quantitative images of the projection data of the atomic number density of each crystalline phase. These images indicate that there is SS400 at the left hand side, SUS304 at the right hand side, and the welded (mixture) line is clearly visualized by the improved RITS code. Thus, it is indicated that the improved RITS code can automatically distinguish the existence of each phase. Furthermore, the quantified values (SS400: $\sim 5.1\times10^{22}$ cm$^{-2}$ and SUS304: $\sim 5.8\times10^{22}$ cm$^{-2}$) correctly correspond to the theoretical values.

Fig. 5 (a-2) and (b-2) show the quantitative images of the degree of texture (crystallographic anisotropy) of each crystalline phase, represented by the March-Dollase (MD) coefficient. The analyses using the RITS code were performed under the theoretical condition that the preferred orientations were $<111>$ along the normal direction of a rolled alpha-iron plate, and $<110>$ along the normal direction of a rolled austenite stainless steel plate. These images indicate that the textures of both alpha-iron and austenite stainless steel are relaxed at the HAZ by the tempering.

Fig. 5 (a-3) and (b-3) show the quantitative images of the crystallite size of each crystalline phase. These images indicate that the crystallite sizes of both phases become larger at the HAZ by the tempering (grain/crystallite growth), as well as the texture. Thus, it is successfully verified that the improved RITS code is applicable for a material composed of multi crystalline phases.
Fig. 3. Photograph of the specimen composed of two crystalline phases. The plate thickness along the $z$ direction is 6 mm. There is alpha-iron (JIS-SS400) having the BCC crystal structure at the left hand side, and is austenite stainless steel (JIS-SUS304) having the FCC structure at the right hand side. There is the welded zone at between them, and is the heat affected zone (HAZ) around the welded zone.
Fig. 4. Experimental data, measured at one pixel, with the fitting curve simulated by the improved RITS code. (a) Only alpha-iron (the BCC phase), (b) only austenite stainless steel (the FCC phase), and (c) both the BCC phase and the FCC phase.
Degree of crystallographic anisotropy of SS400 (BCC) phase (MD coefficient)

Degree of crystallographic anisotropy of SUS304 (FCC) phase (MD coefficient)

Crystallite size of SS400 (BCC) phase (μm)

Crystallite size of SUS304 (FCC) phase (μm)

Projection data of atomic number density of SS400 (BCC) phase (x 10^22 cm^-2)

Projection data of atomic number density of SUS304 (FCC) phase (x 10^22 cm^-2)

Fig. 5. Quantitative imaging of crystalline structural information of a welded two-phases material, obtained by the improved RITS code. Fig. 5 (a) series show the results of the alpha-iron phase, and (b) series show the results of the austenite stainless steel phase. Fig. 5 (1) series show the quantitative crystalline phase images, (2) series show the quantitative crystallographic texture images, and (3) series show the quantitative crystallite block size images, respectively.

5. Conclusion

In this study, we upgraded the RITS code for quantitative evaluation of crystalline structural information included in the transmission spectra obtained by a pulsed neutron imaging experiment. This is because the previous RITS code was not able to deal with all the crystal structures classified into 230 types of space groups and a material composed of multi elements and multi crystalline phases. For improvement of these problems, we implemented some new algorithms for calculation of the crystal structure factor and the total cross section. The demonstrations of simulation calculation and experimental data analysis were successfully carried out. Thus, the improved RITS code is applicable for all the crystal structures of 230 types of space groups, a material composed of multi elements and multi crystalline phases. We are planning to install the concepts of the improved RITS code toward the pulsed neutron imaging instrument at MLF at J-PARC in Japan [18].

Acknowledgements

This work was partially supported by Grant-in-Aid for Scientific Research (S) from Japan Society for the Promotion of Science (No. 23226018).
References

[1] H. Sato, O. Takada, K. Iwase, T. Kamiyama and Y. Kiyanagi: *J. Phys. Conf. Ser.* **251** (2010) 012070.
[2] H. Sato, T. Kamiyama and Y. Kiyanagi: *Mater. Trans.* **52** (2011) 1294.
[3] H. Sato, T. Kamiyama, K. Iwase, T. Ishigaki and Y. Kiyanagi: *Nucl. Instrum. Methods A* **651** (2011) 216.
[4] K. Iwase, H. Sato, S. Harjo, T. Kamiyama, T. Ito, S. Takata, K. Aizawa and Y. Kiyanagi: *J. Appl. Crystallogr.* **45** (2012) 113.
[5] Y. Kiyanagi, H. Sato, T. Kamiyama and T. Shinohara: *J. Phys. Conf. Ser.* **340** (2012) 012010.
[6] J. R. Granada: *Z. Naturforsch.* **39a** (1984) 1160.
[7] G. Placzek: *Phys. Rev.* **93** (1954) 895.
[8] G. Placzek and L. Van Hove: *Il Nuovo Cimento* **1** (1955) 233.
[9] K. Binder: *Phys. Stat. Sol.* **41** (1970) 767.
[10] E. Fermi, W. J. Sturm and R. G. Sachs: *Phys. Rev.* **71** (1947) 589.
[11] S. Vogel: Ph. D. Thesis, Christian Albrechts Universität (2000).
[12] A. C. Larson and R. B. Von Dreele: LAUR 86-748 Los Alamos National Laboratory (2004).
[13] T. M. Sabine, R. B. Von Dreele and J.-E. Jørgensen: *Acta Crystallogr. Sec. A* **44** (1988) 374.
[14] F. Izumi and K. Momma: *Solid State Phenom.* **130** (2007) 15.
[15] K. Iwase, H. Sato, K. Mori, T. Kamiyama, T. Ishigaki and Y. Kiyanagi: *Metall. Mater. Trans. A* **42** (2011) 2296.
[16] K. Mizukami, S. Sato, H. Sagehashi, S. Ohnuma, M. Ooi, H. Iwasa, F. Hiraga, T. Kamiyama and Y. Kiyanagi: *Nucl. Instrum. Methods A* **529** (2004) 310.
[17] H. Sato, O. Takada, S. Satoh, T. Kamiyama and Y. Kiyanagi: *Nucl. Instrum. Methods A* **623** (2010) 597.
[18] Y. Kiyanagi, T. Kamiyama, H. Sato, T. Shinozaka, T. Kai, K. Aizawa, M. Arai, M. Harada, K. Sakai, K. Oikawa, M. Ohi, F. Maekawa, T. Sakai, M. Matsubayashi, M. Segawa and M. Kureta: *Nucl. Instrum. Methods A* **651** (2011) 16.