Residual strain dependence on the matrix structure in RHQ-Nb$_3$Al wires by neutron diffraction measurement

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Abstract

We prepared three types of non-Cu RHQ-Nb$_3$Al wire sample with different matrix structures: an all-Ta matrix, a composite matrix of Nb and Ta with a Ta inter-filament, and an all-Nb matrix. Neutron diffraction patterns of the wire samples were measured at room temperature in the J-PARC 'TAKUMI'. To obtain the residual strains of the materials, we estimated the lattice constant $a$ by multi-peak analysis in the wires. A powder sample of each wire was measured, where the powder was considered to be strain free. The grain size of all the powder samples was below 0.02 mm. For the wire sample with the all-Nb matrix, we also obtained the lattice spacing $d$ by a single-peak analysis. The residual strains of the Nb$_3$Al filament were estimated from the two analysis results and were compared. The resulting residual strains obtained from the multi-peak analysis showed a good accuracy with small standard deviation. The multi-peak analysis results for the residual strains of the Nb$_3$Al filaments in the three samples (without Cu plating) were all tensile residual strain in the axial direction, of 0.12%, 0.12%, and 0.05% for the all-Ta matrix, the composite matrix, and the all-Nb matrix, respectively. The difference in the residual strain of the Nb$_3$Al filament between the composite and all-Nb matrix samples indicates that the type of inter-filament material shows a great effect on the residual strain. In this paper, we report the method of measurement, method of analysis, and results for the residual strain in the three types of non-Cu RHQ-Nb$_3$Al wires.

1. Introduction

Nb$_3$Al has the advantages of better tolerance to strain/stress for stoichiometric composition over Nb$_3$Sn. The rapid-heating, quenching and transformation annealing (RHQ) process enables the formation of stoichiometric Nb$_3$Al with fine grain structures via metastable bcc supersaturated solid solution. As a result a large critical current density of RHQ-Nb$_3$Al is achieved over the whole range of magnetic fields. The RHQ-Nb$_3$Al conductor is very promising for high-field applications such as particle accelerators, fusion reactors, and NMR. The development of RHQ-Nb$_3$Al wires has been reported by Takeuchi et al [1–5]. The critical current $I_c$ of A15-type superconducting wires such as Nb$_3$Al and Nb$_3$Sn is dependent on the tensile strain. Banno et al reported that the $I_c$ of RHQ-Nb$_3$Al wires showed increase and decrease with tensile strain [6].

Since RHQ-Nb$_3$Al wires are composites, thermal phase stress during sample preparation (heat treatment) can be generated due to the differences in the coefficients of thermal expansion (CTEs) among constituent phases. The thermal...
Table 1. Specifications of samples A, B and C.

| Sample           | A    | B    | C    |
|------------------|------|------|------|
| Skin             | Ta   | Nb   | Nb   |
| Inter-filament   | Ta   | Ta   | Nb   |
| Core             | Ta   | Nb   | Nb   |
| Center dummy     | Ta   | Nb   | Nb   |
| Matrix/Nb$_3$Al ratio | 0.8  | 0.8  | 0.8  |
| Number of Nb$_3$Al filaments | 222  | 222  | 144  |
| Diameter of Nb$_3$Al filaments (mm) | 0.04 | 0.04 | 0.05 |
| Outer diameter (mm) | 0.7  | 0.7  | 0.7  |

The cross section of sample A. The skin, inter-filaments, core, and center dummy are all made of Ta.

2. Experimental details

2.1. Sample preparation

We prepared three types of RHQ-Nb$_3$Al wire sample without Cu stabilizer and with different matrix materials (samples A, B and C). The main parameters are summarized as shown in table 1. The cross section of sample A is shown in figure 1.

The following is the sample preparation process. Firstly, a monofilament was prepared by the jelly-roll method [2].



Fig 1

2.2. Sample measurements

The ‘TAKUMI’, a time-of-flight (TOF) neutron diffractometer at MLF/J-PARC was chosen to conduct this study [11, 12]. Multiple peaks in the diffraction pattern were measured simultaneously using two fixed detectors placed at two opposite banks whose diffraction angles with respect to the incident beam were fixed to be $-90^\circ$ and $90^\circ$. Therefore, the diffraction patterns for the wire in the transverse and axial lattice directions could be obtained simultaneously for a beam incident angle of 45°, as shown in figure 2.

For neutron diffraction measurement, seven wires of 20 mm length were bundled. The measurements were performed at room temperature, and the time for a single measurement was 20 min. To confirm the efficacy of the multi-peak analysis by comparing with the single-peak analysis, sample C was measured four times under the same measurement conditions. There might be slight differences in position among the four measurements because multiple samples were set on the goniometer and they were scanned through the measurement. Measurements for the powder samples in sealed cylindrical aluminum holders were carried out by using a radial collimator of 2 mm width [13]. The bore and outer diameters of all the aluminum holders were

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Figure 2. 2D diagram of neutron measurement of wire sample along the transverse and axial directions with two detectors in J-PARC TAKUMI.

8 and 10 mm, respectively. Calibration of TOF and lattice spacing $d$ was performed using CeO$_2$ powder as a standard sample certificated by NIST, and the conversion parameters were determined. Because the volume fraction of Nb$_3$Al was small in all the samples, we used the high intensity mode with a sample-slit width of 5 mm for the incident neutron beam. The experiments were conducted at a proton beam power of 120 kW.

3. Results and discussion

3.1. Neutron diffraction patterns

Neutron diffraction patterns of the wire samples are shown in figure 3. The powder neutron diffraction simulations of Nb$_3$Al, Ta, and Nb were calculated by using Rietan 2000 [14]. In all the patterns, the Nb$_3$Al and matrix material (Ta or Nb) phases were observed without any impurity phase. All the samples showed a difference in the relative intensity ratio of each $hkl$ peak between the transverse and axial directions. This difference indicates that the crystallites in the RHQ-Nb$_3$Al wires have preferred orientation.

3.2. Multi-peak analysis and results

To obtain the residual strain, we firstly determined the lattice constant by a multi-peak analysis for a specific phase in the neutron diffraction pattern. In this study, we applied the multi-peak analysis to the composite wires. The lattice constant $a_m$ of the wire determined by the multi-peak analysis was fitted by using Z-Rietveld (ver. 0.9.34) for more than 10 peaks [16]. The Z-Rietveld was developed for Rietveld analysis of powder samples, but it can also be fitted to non-powder models by setting each peak independently.

Figure 3. Neutron diffraction patterns of wire samples with powder simulations of Nb$_3$Al, Ta, and Nb. The Miller indices of peaks for Nb$_3$Al and Ta (in parentheses) were shown in the axial pattern of sample A.

From the diffraction patterns in the axial and transverse directions in figure 3, it is clearly shown that the wire samples have strong textures. Therefore, the intensity of each peak for a phase was fitted as a free fitting parameter without using a random model or any preferred orientation function. This is different from Rietveld analysis.

The cubic crystal structure of Nb$_3$Al can be deformed to non-cubic structures under anisotropic lattice strain in the composite wire. To estimate the lattice constant $a_m$, the lattice spacings $d$ of each $hkl$ peak were fitted by the linear least-square method that was simulated for the lattice space group $Pm\bar{3}n$ of the powder. Therefore, this is statistical analysis by unification of lattice structures.

The powder samples were also analyzed by multi-peak analysis. The peak intensities were also fitted with independence for each peak. The lattice constant for the powder sample was defined as $a_0$.

As an example, the analysis results for sample A comprised an all-Ta matrix in the transverse and axial directions are shown in figures 4 and 5, respectively. The differences in intensities between the measured and fitted patterns (deltas) have small scatters, indicating a good fitting. The fitted peak positions of Nb$_3$Al and Ta are shown in the figures by vertical lines.

Table 2 shows the lattice constants $a_m$ and $a_0$ for the wire and powder samples, respectively. All the lattice constants $a_m$ were larger than the corresponding lattice constants $a_0$. This indicates that the Nb$_3$Al lattice was extended along the transverse and axial directions in all the wire samples.
3.3. Comparison with single-peak analysis

In order to confirm the efficacy of multi-peak analysis, a comparison with single-peak analysis was carried out. The lattice spacing $d$ for the 211, 320 and 321 peaks of Nb$_3$Al in sample C and its powder was estimated in the axial direction with Gaussian fitting. In the single-peak analysis, we defined the lattice spacings of the wire and powder samples as $d_m$ and $d_0$, respectively. By considering the powder samples to be strain free, the residual strain $\epsilon$ can be evaluated by

$$\epsilon = \frac{d_m - d_0}{d_0}.$$  

For the four measurements on sample C, the results for residual strain obtained by the single-peak and multi-peak analyses are shown in figure 6. The dashed line shows the averaged residual strains of the four measurements for the 211 peak, 320 peak, 321 peak, and multi-peak to be 0.051%, 0.053%, 0.051%, and 0.045%, respectively. The variations in the single-peak analysis results indicate that the residual strain depends on the $hk0$ peaks; it may be related to the diffraction elastic stiffness (to discriminate between the general elastic stiffness of a material and the elastic stiffness obtained by neutron diffraction, we call the latter diffraction elastic stiffness) between the diffraction planes [15]. Considering these variations in the $hk0$ peaks, the residual strains found by the multi-peak analysis method show weighted-averaged values which may express macroscopic residual strains. The standard deviation of the four residual strains obtained from

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**Table 2.** Lattice constants $a_m$ and $a_0$ of the materials for the three samples. The $a_m$ and $a_0$ are the lattice constants of the wire and powder samples, respectively, obtained by the multi-peak analysis.

| Sample | Material | Direction | $a_m$ (Å)   | $a_0$ (Å)   |
|--------|----------|-----------|-------------|-------------|
| A      | Nb$_3$Al | Transverse | 5.1877 ± 0.0001 | 5.1852 ± 0.0005 |
|        |          | Axial     | 5.1919 ± 0.0001 | 5.3027 ± 0.0001 |
|        | Ta       | Transverse | 3.2997 ± 0.0001 | 3.3016 ± 0.0005 |
|        |          | Axial     | 5.1919 ± 0.0002 | 5.1856 ± 0.0005 |
| B      | Nb$_3$Al | Transverse | 5.1867 ± 0.0001 | 5.1860 ± 0.0004 |
|        |          | Axial     | 5.1887 ± 0.0001 | 5.1891 ± 0.0001 |
|        | Nb       | Transverse | 3.3004 ± 0.0001 | 3.3019 ± 0.0006 |
|        |          | Axial     | 3.2991 ± 0.0001 | 3.3000 ± 0.0001 |

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**Table 3.** CTEs and Young’s moduli of the materials at room temperature [7, 8].

| Material | CTE ($10^{-6}$ K$^{-1}$) | Young’s modulus (GPa) | Poisson’s ratio |
|----------|--------------------------|-----------------------|-----------------|
| Ta       | 6.4                      | 185                   | 0.34            |
| Nb       | 7.1                      | 103                   | 0.40            |
| Nb$_3$Al | 8.75                     | 169                   | 0.3             |
| Nb$_3$Sn | 7.2                      | 165                   | 0.3             |
Figure 6. Residual strains of the Nb\textsubscript{3}Al filament in sample C from single-peak and multi-peak analyses in the axial direction. The dashed line shows the average of the four measurements. The circular arrow line shows the error by the standard deviation this average. In the multi-peak analysis, the strain was calculated from the lattice constant $a$ that was analyzed by using Z-Rietveld. The multi-peak analysis is 0.001%. This value is 3–9 times smaller than that obtained from single-peak analysis. This indicates that the statistics by multi-peak analysis in wire certainly is valid and feasible to improve the accuracy.

3.4. Residual strains in wires with different matrices

An elastic behavior with isotropic thermal strain is assumed in the samples. For samples A, B and C, the residual strains of the Nb\textsubscript{3}Al filament obtained from the multi-peak analysis are shown in figure 7 with the corresponding cross sections of the wires. Considering the small systematic errors such as in the sample position and clock time (TOF), the error in the residual strain caused by the systematic errors is below 0.01%. The values of the error bars were obtained by summation of the error caused by the systematic errors and the error calculated by using the errors of the lattice constants $a$ from Z-Rietveld. All the residual strains had tensile strain in the axial direction. In all the samples, the residual strains of the Nb\textsubscript{3}Al filament in the axial direction were larger than those in the transverse direction. This indicates that the thermal stress shows a significant effect in the axial direction. Similar behavior can be seen in the strain distribution of Nb\textsubscript{3}Sn [10].

In the given direction, samples A and B showed almost the same residual strain in the Nb\textsubscript{3}Al filament. This result indicates that the residual strain of the Nb\textsubscript{3}Al filament is dominantly influenced by the adjacent inter-filament matrix of Ta and the effects of the skin, core, and center dummy are relatively small. The residual strain of the Nb\textsubscript{3}Al filament in sample B was larger than that in sample C by about 0.08% and 0.02% in the axial and transverse directions, respectively. The residual strain of the Nb\textsubscript{3}Al filament has a great dependence on the materials of the inter-filament.

We also estimated the residual strain of the matrices of Ta and Nb by the multi-peak analysis. The crystal structures of Ta and Nb have the same space group $P_{m\overline{3}}$ and almost the same parameters. Thus, it is difficult to distinguish the Ta and Nb peaks in the diffraction patterns of sample B in which both Ta and Nb are utilized for the matrix material. The analysis results for the matrix materials in samples A and C are shown in figure 7. In the axial direction, the Ta and Nb show compressive residual strain corresponding to the tensile strain of the Nb\textsubscript{3}Al filament.

Figure 7. Residual strains obtained from the multi-peak analysis. The residual strains in samples A and C were calculated in the axial direction as shown in the model because of thermal contraction.
3.5. Model calculation of the residual strain induced by thermal contraction

We calculated the residual strains of the Nb$_3$Al filament and matrix materials by a thermal contraction model for samples A and C in the axial direction. In this model, the transverse strains were assumed to have no effect on the axial strains. During heat treatment, as the temperature decreased below 800°C, axial residual strain was assumed to be generated at 550°C ($T_H$) [15, 17]. Then, as the temperature decreased to room temperature ($T_R$), the residual strain increased with an increase in the stress between the Nb$_3$Al filament and the matrix material owing to the difference in their CTEs.

The CTE of Nb$_3$Al is larger than that of Ta or Nb; therefore the Nb$_3$Al filament has tensile residual strain in the axial direction. The model representation of thermal contraction is shown in figure 8. In figure 8(a), the axial length of the wire is defined as $L_0$, at 550°C when the wire is free of residual strain. The axial lengths of the Nb$_3$Al filament and the matrix material at room temperature are $L_a$ and $L_b$, respectively, considering their thermal contraction separately as shown in figures 8(b) and (c). In actuality, the composite wire shrank to length $L_1$ with residual strains between the Nb$_3$Al filament and the matrix material at room temperature, as shown in figure 8(d). The residual strains of the Nb$_3$Al filament and the matrix materials in the axial direction were calculated as follows.

At room temperature, the stresses $\sigma$ between the Nb$_3$Al filament and the matrix material in the axial direction are given as

$$\int_0^{r_a} \sigma_a \, dr + \int_{r_a}^{r_b} \sigma_b \, dr = 0. \quad (1)$$

The thermal strains $\epsilon'$ are expressed as

$$\epsilon'_a = \int_{T_H}^{T_R} (-C_a) \, dT = \frac{L_a - L_0}{L_0} \quad (2)$$

$$\epsilon'_b = \int_{T_H}^{T_R} (-C_b) \, dT = \frac{L_b - L_0}{L_0}. \quad (3)$$

The residual strains are

$$\epsilon_a = \frac{(E_{af} + E_{bf})(1 + \epsilon'_b)}{E_{af}(1 + \epsilon'_a) + E_{bf}(1 + \epsilon'_b)} - 1 \quad (4)$$

$$\epsilon_b = \frac{(E_{af} + E_{bf})(1 + \epsilon'_a)}{E_{af}(1 + \epsilon'_a) + E_{bf}(1 + \epsilon'_b)} - 1 \quad (5)$$

where $E$ is the Young’s modulus, $f$ is the volume fraction, and $\epsilon$ is the residual strain between the Nb$_3$Al filament and the matrix material.

The calculation parameters for Nb$_3$Al, Ta, and Nb at room temperature are shown in tables 1 and 3.

The calculation results for residual strain are shown in figure 7. In sample A, the residual strain obtained by the model calculation was less than that obtained by the multi-peak analysis for the Nb$_3$Al filament and the former was larger than the latter for Ta. This indicates that the strain mechanism for the all-Ta matrix is different from that for the all-Nb matrix in the axial direction. Considering the different strain mechanisms of samples A and C in the transverse direction, this may be related to the effect from the transverse direction due to the high hardness of Ta and the complicated structure of the cross section.

In sample C, the calculation results for the Nb$_3$Al filament and Nb were similar to the results obtained by multi-peak analysis. With the model calculation, the residual strain of the Nb$_3$Al filament for sample C at 4.2 K was 0.07%. Also, the residual strain of the Nb$_3$Al filament with Cu stabilizer at room temperature was calculated by the model calculation as 0.01%. It was considered that effective thermal stress appears between the Cu stabilizer and the matrix from 300°C in the cooling process with the recovery temperature of Cu. The calculation result shows that the residual strain of the Nb$_3$Al filament has a strong effect from the Cu stabilizer due to the large CTE of Cu. Since the yield strain of Cu is small, it is difficult to estimate the residual strain quantitatively. However, considering the strong compressive effect of the Cu stabilizer on the matrix, the residual strain of the Nb$_3$Al filament is estimated to be compressive at 4.2 K.

4. Conclusion

The residual strains for the Nb$_3$Al filament and matrix materials in RHQ-Nb$_3$Al wires were measured by neutron diffraction at room temperature. We estimated the lattice constants $a_m$ for three types of wire by multi-peak analysis with anisotropic lattice strains in the axial and transverse
directions of the wire, due to the different diffraction elastic stiffnesses, and sample configurations. The multi-peak analysis showed good accuracy in the estimation of the residual strain in RHQ-Nb$_3$Al wires. We estimated the tensile residual strain of the Nb$_3$Al filament and the compressive strain of the Ta and Nb in the axial direction, by analyzing the lattice constant $a_m$. By comparing the all-Nb matrix and the composite matrix samples, the residual strain is greatly affected by the type of inter-filament material. We also obtained the difference in the residual strains of the Nb$_3$Al filament between the all-Ta matrix and all-Nb matrix samples in the RHQ-Nb$_3$Al wires, and the residual strains were calculated by the thermal strain model in the axial direction. The result was that the all-Ta matrix and all-Nb matrix samples showed different strain mechanisms in the axial and transverse directions.

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