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In Situ Neutron Diffraction during Martensitic Transformation by Subzero Cooling in an Fe-33Ni Alloy

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An austenitic single phase Fe-33Ni alloy was cooled down to 120 K and warmed up to RT, and subsequently cooled to 4 K followed by re-heating to RT. The athermal martensitic transformation occurred below 190 K. During the heat cycles, *in situ* neutron diffraction experiments with a TOF method were carried out at KENS/KEK. The changes in lattice plane strain during the martensitic transformation in a bulk specimen were obtained from the profile analysis. The averaged internal stress in the austenite yielded by the transformation has been estimated.

KEYWORDS: martensitic transformation, internal stress, neutron diffraction, time of flight method, Fe-Ni alloy

§1. Introduction

The martensitic transformation is diffusionless and accompanies shape change, i.e., transformation strains which generate the internal stresses.[1)] Lattice defects are introduced as a result of plastic relaxation of locally high internal stresses in most cases. To control the internal stresses and lattice defects is a very important issue of engineering applications, for instance, strengthening of steels, development of shape memory alloys and improvement of ductility in ceramics. Such an internal stress evolution has, however, never been measured directly so far. This may presumably be because X-ray diffraction, which can easily be employed, is not appropriate to obtain bulky data. Recently, neutron diffraction methods for determination of internal stresses in structural materials have found lots of applications in the field of materials science and engineering because of a high penetration ability of thermal neutrons.[2–5)] Then, in this study, neutron diffraction experiment for an austenitic single phase Fe-33Ni alloy was performed. A bulk specimen was cooled to a cryogenic temperature and then heated up to RT. During the heat cycle, *in situ* neutron diffraction experiments with a TOF method were carried out at KENS/KEK. The changes in lattice interplanar spacings during austenite (γ: fcc) to martensite (α': bcc) athermal transformation were measured. The results obtained from a profile analysis are discussed in comparison with theoretical considerations.

§2. Experimental Procedures

An alloy used in this investigation was prepared by vacuum induction melting. An obtained ingot was homogeneized and hot-forged to a plate. The results of chemical analysis are 32.85Ni, 0.004C, <0.01Mn, <0.01Si, <0.004P, 0.007S and balanced Fe (in mass%). Round bar specimens with a diameter of 6 mm and a length of 50 mm were made and austenitized at 1423 K for 10.8 ks followed by furnace cooling.

The specimen was inserted into a vanadium tube that was filled with helium gas. The tube was set in a specimen chamber of a TOF neutron diffractometer, VEGA,[6)] at KENS/KEK. Two thermal cycles were given to the specimen, during which diffraction profiles were obtained at various temperatures. Temperature was controlled by using a cryo-cooler and a heater within a deviation of 2 K. In the first cycle, the temperature was lowered to 120 K and then warmed up to RT (296 K). Subsequently, in the second run, the specimen was cooled down to 4 K followed by warming to RT. Diffraction profiles were obtained for approximately 3.6 ks at each measuring temperature.

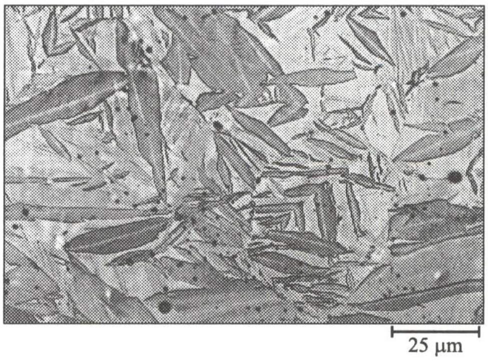

Fig. 1. Martensite (α') grains formed in the austenite (γ) matrix observed by a light microscope: a specimen was cooled to 77 K followed by warming up to RT and then etched by a 5% nital solution.

The (hkl) lattice plane strains in the austenite matrix were determined from the deviations of lattice interplanar spacings (d_{hkl}) from the stress-free state. This was possible for γ while only the change in d_{hkl} was obtained in α'. Once α' was introduced, the stress-free spacing cannot be obtained even for γ. Lattice plane strain ϵ_{hkl} is calculated from the displacement of the profile center, which is measured as wavelength difference ($\Delta\lambda$), i.e., flight-time difference (Δt) in the TOF method.

$$\epsilon_{hkl} = (d_{hkl} - d^0_{hkl})/d^0_{hkl} = \Delta\lambda/\lambda = \Delta t/t \qquad (2.1)$$

where λ and t are the neutron wavelength and the flight-time for (hkl) plane at the stress-free state, respectively.

§3. Experimental Results and Discussions

3.1 Martensitic transformation by subzero-cooling

The specimens showed fully austenitic single phase at RT after the solution treatment. From differential scanning calorimetry (DSC) experiments, the Ms temperature was identified to be 190 K. Figure 1 shows an example of athermally induced α' grains which are lenticular martensite with a midrib.

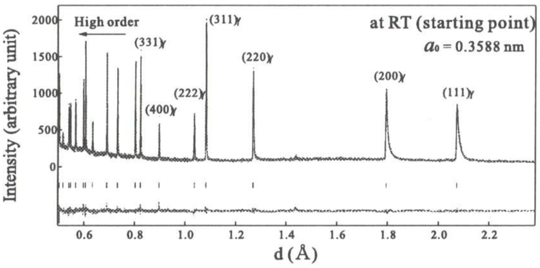

Fig. 2. Neutron diffraction profile obtained at RT (before cooling: single γ phase).

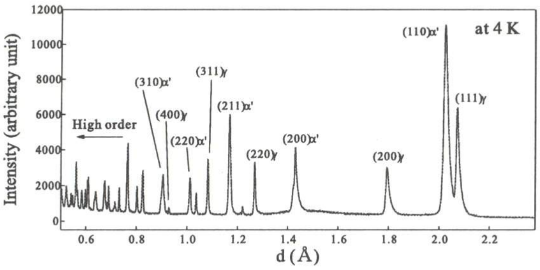

Fig. 3. Neutron diffraction profile obtained at 4 K (α'-γ mixture).

Examples of diffraction profiles obtained are shown in Figs.2 and 3. At RT as the starting point, the specimen was found to consist of only γ as recognized in Fig.2. When the Rietveld refinement was applied to the diffraction profile, the lattice parameter of γ at RT was determined to be 0.3588 nm. As seen in the figure, the fitting was excellent (see the residuals plotted at the bottom).

On the other hand, at 4 K in Fig.3, the formation of α' can be found. The Rietveld refinement may be applicable only for the single γ structure, because an α'-γ mixture holds internal stresses and the elastic deformation behavior and thermal expansion behavior show orientation dependences. Hence, from here on, the profile analysis was performed for individual single peaks.

3.2 Change in martensite volume fraction during cooling and heating

Figure 4 shows the intensity ratio of (211)α' to (200)γ as a function of temperature. As can be seen, the volume fraction of α' increases with lowering the cooling temperature at the first cycle. The increase in the ratio starts between 180 and 200 K, which is almost consistent with the Ms temperature determined by the DSC test. Because the As temperature was higher than RT, the reverse transformation did not take place during the heating to RT. When we employed a conventional X-ray method for estimation of α' volume fraction from the diffraction intensity ratio, it was approximately 50% at 120 K.

At the second cycle down to 4 K, a little more amounts of α' were produced. Noted here was the fact that γ was still retained even at 4 K.

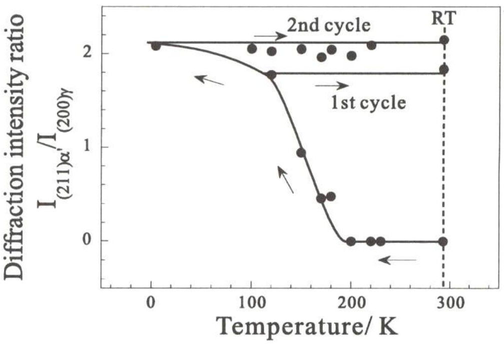

Fig. 4. Changes in the ratio of diffraction peak heights (211)α' to (200)γ.

3.3 Lattice plane strain changes in austenite by cooling and heating

In order to convert the lattice interplanar spacing to strain, the reference spacing of the stress-free state is necessary. Such a reference is available only for γ and the value at the starting point (RT) was employed here. Then, the strain in γ was calculated and the results obtained from (200)γ for the first cycle was shown in Fig.5.

In the beginning, the (200) lattice plane strain in γ decreases with thermal contraction and then increase markedly relating to the athermal martensitic transformation, i.e., transformation strains (expansion). During the heating from 120 K to RT, the lattice plane strain increased due to thermal expansion of an α'-γ mixture, whose thermal expansion coefficient is lower than that of γ.

The obtained strains stem from three origins: (1) thermal contraction or expansion, (2) thermal misfit strain due to different thermal coefficients of γ and α' and (3) transformation strains.

3.4 Estimation of internal stress caused by the transformation strain

The γ to α' martensitic transformation in an Fe-high Ni alloy has been known to accompany shape changes of shear and dilatation. The transformation strains, i.e., misfit strains are complemented by elastic strain (internal stress) and plastic flow within an α' grain itself and in the γ matrix. According to Breedis and Wayman's work for an Fe-30.9 mass% Ni alloy,[7] the lattice constants for γ and α' were reported as 0.3591 nm (almost identical with the present result) and 0.2875 nm, respectively. Although the shear and dilatation directions in an individual α' grain are strictly related to the K-S orientation relationship, the averaged transformation strains in a non-textured polycrystal are regarded as isotropic, i.e., simple volume expansion. That is, the volume strain calculated from the above reported lattice constants is 2.63%. Thus, let us assume the transformation strains ϵ^T_{ij} in the present Fe-33 mass% Ni alloy as,

$$\epsilon^T_{ij} = \epsilon^T \delta_{ij}, \qquad (3.1)$$

where δ_{ij} is Kroneker's delta. Here ϵ^T was taken as 0.878% that was estimated from the volume strain reported for the Fe-30.9mass% Ni alloy. The internal stresses yielded in the both component phases are also considered to be of an iso-static condition.

Based on the above assumptions, the average internal stress in γ could be predicted by using the Eshelby[8] and the Mori-Tanaka[9] theories. Similarly to the case of misfit strain due to different thermal expansions in two component phases,[4] the average internal stress in γ (σ^{γ}_{ij}) can be obtained by

$$\sigma^{\gamma}_{ij} = -fC_{ijkl}(S_{klmn} - I)\epsilon_{mn}, \qquad (3.2)$$

where *f*: volume fraction of α', C_{ijkl}: elastic moduli, S_{klmn}: Eshelby's tensor ($S_{1111} = S_{2222} = S_{3333} = (7 - 5\nu)/15(1-\nu), S_{2233} = S_{3311} = S_{1122} = S_{3322} = S_{1133} = S_{2211} = (5\nu - 1)/15(1-\nu), S_{1212} = S_{2323} = S_{3131} = -(4-5\nu)/15(1-\nu)$ for sphere) and *I*: identity matrix. Here, the elastic moduli of γ and α' are assumed to be identical for simplicity.

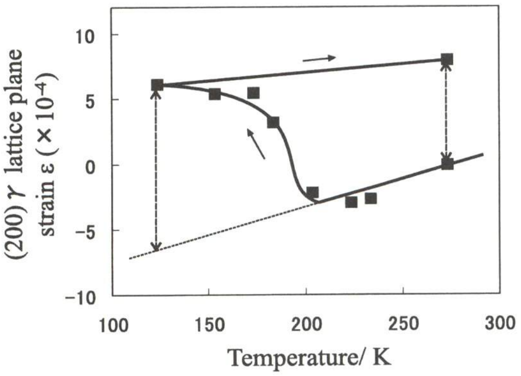

Fig.5. Changes in (200)γ lattice plane strain during the first cooling and heating cycle.

When we input the Young modulus $E = 200$ GPa and $\nu = 0.3$, we obtain

$$\epsilon_{11} = \epsilon_{22} = \epsilon_{33} = 0.00336f \qquad (3.3)$$

as the average elastic strain in γ. The averaged internal stress in γ is given by

$$\sigma_{11} = \sigma_{22} = \sigma_{33} = 1680f(MPa) \qquad (3.4)$$

When *f* is 0.50 as estimated at 120 K in the first cycle in section 3.3, ϵ_{11} ($= \epsilon_{22} = \epsilon_{33}$) becomes 0.00168. For comparison with the predictions and the measurements, to be noted is that thermal contraction or expansion must be taken into account.Then, if we extrapolated the thermal contraction line for γ in Fig.5 (see dashed line), the lattice plane strain would be approximately 0.0013. This is roughly in a good agreement with the above prediction. The average internal stress in γ is thus predicted to be approximately 840 MPa. This stress would decrease when the specimen was warmed up to RT because the misfit thermal strain between α' and γ has an opposite sign compared with the transformation strain.

§4. Conclusions

The internal elastic strains in the austenite matrix caused by martensitic transformation on cooling were measured in a bulky specimen by means of the TOF in situ neutron diffraction. The round bar specimen, which showed austenitic single phase at RT, was cooled down to a cryogenic temperature followed by heating up to RT. During the cooling, athermal martensitic transformation occurred resulting in an internal stress state. The diffraction profiles were obtained at various temperatures during the cooling and heating. From the results of a profile analysis, the lattice plane spacings were found to change as a function of temperature. The changes are caused by (1) thermal contraction and expansion, (2) thermal phase stress due to misfit thermal strains between the constituents, and (3) internal stress due to the transformation strain. The internal stress in the austenite yielded by martenisitc transfomation has been estimated about 840 MPa in a hydrostatic stress condition when the volume fraction of the martensite formed was 50%.

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