Pseudoelastic Behavior of Fe–Al Polycrystals

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Pseudoelastic behavior of Fe–Al polycrystals at room temperature was examined focusing on Al concentration, the crystallographic texture and the grain size. Perfect pseudoelasticity derived from the reversible motion of $1/4\langle 111 \rangle$ superpartial dislocations dragging the anti-phase boundaries (APB) took place in Fe–25.0at%Al polycrystals at a total strain of 1.0%. The amount of strain recovery in Fe–Al polycrystals showed a maximum at 25.0at% Al and the deviation from the concentration led to a decrease in strain recovery. The backward stress due to APB and dislocation configuration in Fe–Al alloys was closely related to the dependence of the pseudoelasticity on Al concentration. The pseudoelastic behavior of Fe–25.0at%Al polycrystals depended strongly on the loading axis, which could be accounted for in terms of the Taylor factor. Moreover, recovery strain of Fe–23.0Al polycrystals increased with increasing average grain size suggesting that the grain boundaries suppressed the forward and backward motion of the superpartials and were harmful for the pseudoelasticity.

KEY WORDS: Fe₃Al; intermetallic compounds; dislocations; crystallographic texture; grain size.

1. Introduction

Shape memory alloys are known to exhibit shape memory effect and pseudoelasticity by which an applied strain recovers during heating and unloading, respectively.¹⁾ In general, pseudoelasticity originates from a stress-induced martensitic transformation during loading and the reverse transformation during unloading, respectively. However, Fe–23.0Al (at%) single crystals with the D0₃ structure (Fe₃Al lattice) demonstrated perfect pseudoelasticity at room temperature regardless of the martensitic transformation.^{2–4)} It is interesting to note that the pseudoelasticity of Fe–Al alloys was based on the peculiar dislocation motion in the D0₃ lattice. The $\langle 111 \rangle$ superdislocation was generally dissociated into four $1/4\langle 111 \rangle$ superpartial dislocations bound by the nearest-neighbor and the next-nearest-neighbor antiphase boundaries (NNAPB and NNNAPB). In Fe–23.0Al single crystals, the leading $1/4\langle 111 \rangle$ superpartials moved individually dragging the NNAPB during loading. During unloading, the NNAPB pulled back the superpartials due to its tension resulting in the pseudoelasticity. Therefore, we call this phenomenon “APB pseudoelasticity”.

In general, mechanical properties of polycrystalline materials depend strongly on the microstructure such as grain size and crystallographic texture.⁵⁾ For instance, a yield stress of polycrystals increases with decreasing grain size through the Hall–Petch law since grain boundaries usually suppress the dislocation motion. Therefore, mechanical properties of polycrystals are generally different from those of the single crystals. Although the pseudoelastic behavior of Fe₃Al single crystals was extensively investigated, no

systematic study using the polycrystals has not been carried out yet. In this paper, we report the pseudoelastic behavior of Fe–Al polycrystals focusing on Al concentration, the crystallographic texture and the grain size.

2. Experimental Procedure

Master ingots of Fe–16.0~28.0Al (at%) were prepared by melting high purity Fe and Al in a plasma arc furnace in a purified Ar atmosphere. The ingots 12 mm in thickness were hot-rolled to 70% reduction at 900°C and then warm-rolled to 80% at 600°C. After the rolling, the specimens were annealed at 700–1 000°C for 1 h for recrystallization followed by slow cooling to room temperature at a cooling rate of 100°C/h for the D0₃ ordering. The microstructure of Fe–Al alloys was well documented in our previous paper.⁴⁾ At and above 23 at% Al, the D0₃ single phase developed and there existed numerous ordered domains. In contrast, the (bcc+D0₃) two-phase microstructure or short-range order was observed below 23 at% Al. The tensile specimens of which gauge dimension was 1.5×0.7×5.0 mm³, were cut from the recrystallized sheets by spark machining. The angle between loading axis and the rolling direction (θ) was set to be 0°, 45° and 90°. Cyclic tensile tests with incremental strain amplitudes (ϵ_t) were performed in air at room temperature at constant cross-head speed of 0.05 mm/min during loading and unloading. The applied strain amplitude was measured by a charge-coupled devices and was increased 1.0% per cycle. Note that the cross-head speed corresponded to an initial strain rate of 1.7×10^{-4} /s. The amount of strain recovery was evaluated using recovery ratio (R) defined as follows:

$$R = \frac{\varepsilon_t - \varepsilon_r}{\varepsilon_t} \times 100 \dots\dots\dots(1)$$

where ε_t and ε_r are the total strain and the residual strain, respectively. Microstructure of the specimens was analyzed by an electron back-scatter diffraction (EBSD) method. The electron beam was automatically moved in 10 μm steps to develop an orientation map (OIM).

3. Results

3.1. Microstructure of Fe–Al Polycrystals

Figure 1 shows OIM maps of Fe–23.0Al warm-rolled sheets before and after recrystallization at 800°C for 1 h. In as warm-rolled specimens, there exists two types of deformation bands oriented $\langle 111 \rangle$ and $\langle 110 \rangle$ directions parallel to the normal and rolling directions (ND and RD), respectively (Figs. 1 (a), 1(b)). This means the α (RD// $\langle 110 \rangle$) and γ (ND// $\langle 111 \rangle$) fiber textures develop in the as warm-rolled Fe–23.0Al sheets. After the recrystallization treatment at 800°C, the frequency of grains belonging to the γ fiber texture increases as shown in Figs. 1 (c) and 1(d). It is also noted that the equiaxed grains of which the average grain size ($\bar{d}$) is 43.6 μm can be observed after annealing at 800°C. Such rolling and recrystallization textures are similar to those of bcc metals.⁵⁾ **Figure 2** shows orientation distribution function (ODF) at $\phi_2=45^\circ$ for Fe–16.0~28.0Al polycrystals after recrystallization at 800°C for 1 h. Below 23 at% Al, development of near-Cube texture ($\phi_1=45^\circ$, $\Phi=0^\circ$) can be seen as shown in Figs. 2 (a) and 2(b). In contrast, the intensity around $\Phi=55^\circ$ becomes strong at and above 23 at% Al suggesting that the γ fiber texture develops in the Fe–Al sheets after the recrystallization (Figs. 2(c)–2(e)). It is also noted that the γ fiber texture connects to $\{114\}\langle 481 \rangle$ component which generally develops in bcc metals at higher reduction in thickness.⁶⁾ The $\bar{d}$ of Fe–23.0Al polycrystals recrystallized at 700–1 000°C was measured using the EBSD technique. The $\bar{d}$ increased from 26.9 to 140.6 μm with increasing recrystallization temperature from 700 to 1 000°C.

3.2. Pseudoelastic Behavior of Fe–Al Polycrystals

Stress–strain (S–S) curves of Fe–25.0Al polycrystals recrystallized at 800°C for 1 h and then pulled with $\theta=0^\circ$ at room temperature are shown in **Fig. 3**. In the repeated tensile tests, ε_t was increased 1.0% per cycle. At $\varepsilon_t=1.0\%$, perfect pseudoelasticity takes place in the Fe–25.0Al polycrystals. On the other hand, the residual strain becomes large as ε_t increases and the S–S curve shifts to higher strains. Note that the recoverable strain of Fe–23.0Al single crystals was approximately 5.0%, indicating that the pseudoelastic properties of Fe–Al polycrystals were inferior to those of the single crystals. However, in other words, a recoverable strain of 1.0% could be obtained even in the Fe–Al polycrystals. The R values of Fe–Al alloys at $\theta=0^\circ$ and 90° after recrystallization at 800°C for 1 h are plotted against Al concentration in **Fig. 4**. The R values at $\varepsilon_t=1.0$ and 2.0% show a maximum at 25.0 at% Al both at $\theta=0^\circ$ and 90° (Figs. 4(a) and 4(b)) and decreases with increasing the deviation from the stoichiometric concentration. In the

Fig. 1. OIM of Fe–23.0Al warm-rolled sheets before (a), (b) and after recrystallization (c), (d) at 800°C for 1 h. The grain color corresponds to the unit stereo triangle shown in (e). (a), (c) OIM for ND, (b), (d) OIM for RD.

Fig. 2. ODF section at $\phi_2=45^\circ$ for Fe–16.0~28.0Al polycrystals after recrystallization at 800°C for 1 h. (a) Fe–16.0Al, (b) Fe–20.0Al, (c) Fe–23.0Al, (d) Fe–25.0Al and (e) Fe–28.0Al. Ideal positions for each texture component are plotted in (f).

Fig. 3. S–S curves of Fe–25.0Al polycrystals recrystallized at 800°C for 1 h and then pulled to different ε_t with $\theta=0^\circ$ at room temperature.

Fig. 4. Dependence of R on Al concentration in Fe–Al polycrystals recrystallized at 800°C for 1 h and then tensile deformed to $\varepsilon_t = 1.0$ and 2.0% with $\theta = 0^\circ$ (a) and 90° (b) at room temperature.

Fig. 5. Changes in R and σ_y with θ in Fe–25.0Al polycrystals recrystallized at 800°C for 1 h and then tensile deformed to $\varepsilon_t = 1.0$, 2.0 and 3.0% at room temperature.

case of Fe–Al single crystals, however, the optimum Al concentration of the pseudoelasticity was 23 at%, different from that in the polycrystals.⁴⁾

The R value of Fe–25.0Al polycrystals recrystallized at 800°C for 1 h and then tensile deformed to $\varepsilon_t = 1.0$, 2.0 and 3.0% depends strongly on θ , as shown in Fig. 5. The R value at $\theta = 0^\circ$ and 90° is much higher than that at $\theta = 45^\circ$. The anisotropy of the R value depending on θ decreases with increasing ε . The yield stress (σ_y) of the polycrystals is also plotted against θ , showing opposite tendency to the R value.

Figure 6 shows relationship between $\bar{d}$ and R value in Fe–23.0Al polycrystals tensile deformed to $\varepsilon_t = 3.0\%$ at $\theta = 0^\circ$. Fe–23.0Al polycrystals annealed at 700°C with $\bar{d} = 26.9 \mu\text{m}$ showed small R value. However, the R value increases with increasing $\bar{d}$ and reaches to 45% with $\bar{d} = 140.6 \mu\text{m}$. Thus, the pseudoelasticity of Fe–Al crystals is closely related to $\bar{d}$.

4. Discussion

In the present study, we first found that not only Fe–Al single crystals but also the polycrystals demonstrated large pseudoelasticity. In particular, $\varepsilon_t = 1.0\%$ was recoverable in Fe–25.0Al polycrystals. At present, there was no indication that martensites or twins were responsible for the pseudoelasticity in Fe–Al polycrystals. Thus, it is reasonable to consider that APB pseudoelasticity appeared in the poly-

Fig. 6. Variation in R with $\bar{d}$ in Fe–23.0Al polycrystals pulled to $\varepsilon_t = 3.0\%$ at room temperature; $\theta = 0^\circ$.

crystals as well as the single crystals.

Let us consider the effect of Al concentration on the pseudoelastic behavior of Fe–Al polycrystals focusing on the stress acting on $1/4\langle 111 \rangle$ superpartials. When APB pseudoelasticity appeared in Fe–Al crystals, the NNAPB pulled back $1/4\langle 111 \rangle$ superpartials during unloading, resulting in the pseudoelasticity.^{2–4)} The NNAPB back stress acting on the superpartials is given by

$$\tau_{\text{NN}} = \frac{\gamma_{\text{NN}}}{b} \quad \text{.....(2)}$$

where b is the magnitude of the Burgers vector of $1/4\langle 111 \rangle$ and the NNAPB energy (γ_{NN}) is written as⁷⁾:

$$\gamma_{\text{NN}} = \frac{2\sqrt{2}}{a_0^2} \{4V_1S_1^2 + V_2(S_2^2 - 4S_1^2)\} \quad \text{.....(3)}$$

where a_0 is the lattice constant, V_1 and V_2 is the first- and second-nearest ordering energies and S_1 and S_2 are the first- and second-nearest degrees of order, respectively. It should be noticed that the τ_{NN} obtained from Eqs. (2) and (3) was quantitatively consistent with the shape of stress–strain curve in Fe–Al single crystals; the detail is well documented in our previous papers.^{3,4)} Furthermore, in general, the τ_{NN} was closely related to the pseudoelastic behavior: the higher the τ_{NN} , the higher the R . Therefore, the variation of τ_{NN} with Al content was evaluated as follows. S_1 and S_2 in Eq. (3) correlate with Al concentration, written as⁷⁾:

$$S_1 = 2k_1x \quad (0 \leq x \leq 0.50) \quad \text{.....(4)}$$

$$S_2 = 4k_2x \quad (0 \leq x \leq 0.25) \\ = 4k_2(1-2x) \quad (0.25 \leq x \leq 0.50) \quad \text{.....(5)}$$

where x is the mol fraction of Al, k_1 and k_2 are the constant. If k_1 and k_2 were set to be 0.71 and 0.66, respectively, S_1 and S_2 calculated from Eqs. (4) and (5) agreed well with those measured by X-ray analysis.⁴⁾ By substituting other parameters in Eqs. (2)–(5), the τ_{NN} was calculated and is plotted in Fig. 7. The τ_{NN} increases almost linearly with increasing x up to 0.25 and then remains almost constant by further increase of x to 0.3. At lower Al contents, the backward motion of $1/4\langle 111 \rangle$ superpartials was considered to be difficult because of the low τ_{NN} . This resulted in the decrease in R below $x = 0.23$. On the other hand, higher τ_{NN} at $x = 0.23$ and 0.25 enabled $1/4\langle 111 \rangle$ superpartials to reversibly move during loading and unloading. Consequently,

Fig. 7. Variation in τ_{NN} and τ_{NNN} with Al content in Fe–Al polycrystals.

large pseudoelasticity took place at $x=0.23$ and 0.25 . In contrast, the R above $x=0.25$ exhibited small values in contrast to high τ_{NN} . This is because the dislocation configuration changed above $x=0.25$; paired $1/4\langle 111 \rangle$ superpartials started to move dragging the NNNAPB.^{3,4)} The backward stress of the NNNAPB (τ_{NNN}) is given by⁷⁾

$$\tau_{NNN} = \frac{\gamma_{NNN}}{2b} \dots\dots\dots(6)$$

where γ_{NNN} is the NNNAPB energy written as:

$$\gamma_{NNN} = \frac{2\sqrt{2}}{a_0^2} \{2V_2 S_2^2\} \dots\dots\dots(7)$$

The τ_{NNN} is also shown in Fig. 7. Note that the denominator of Eq. (6) is two times larger than that of Eq. (2) while the γ_{NNN} shows similar value to the γ_{NN} at around $x=0.25$. As a result, the τ_{NNN} is much smaller than the τ_{NN} , resulting in small R above $x=0.25$. Therefore, the dependence of pseudoelastic behavior in Fe–Al polycrystals on Al concentration could be interpreted in terms of the backward stress originating from the NNAPB and the dislocation configuration.

It should be noted that Fe–Al single crystals demonstrated maximum R at 23.0 at% Al while the optimum Al concentration for the pseudoelasticity in the polycrystals was 25.0 at% Al.^{3,4)} In Fe–Al single crystals, the dislocation configuration of $1/4\langle 111 \rangle$ superpartials strongly affected the pseudoelastic behavior. The individual motion of $1/4\langle 111 \rangle$ superpartials dragging the NNAPB resulted from the interaction between the superpartials and the boundaries of the DO_3 ordered domains.^{3,4)} The fine ordered domains less than 50 nm in diameter, developed at 23.0 at% Al were optimum for $1/4\langle 111 \rangle$ superpartials moving independently, resulting in the maximum R at the Al content in the single crystals. On the other hand, the domain size increased with increasing Al content and consequently, the activation of paired $1/4\langle 111 \rangle$ superpartials dragging the NNNAPB led to the decrease in R .^{3,4)} In Fe–Al polycrystals, however, the grain boundaries suppressed the motion of $1/4\langle 111 \rangle$ superpartials. Furthermore, several slip systems were necessarily activated in the polycrystals to transfer strain across the grain boundaries. The interaction between the activated slip systems resulted in the formation of sessile dislocations. Consequently, the frictional stress of $1/4\langle 111 \rangle$ superpartials

in Fe–Al polycrystals became larger than that of the single crystals. Under such circumstances, higher τ_{NN} was required for the backward motion of the superpartials in Fe–Al polycrystals, compared with the single crystals. The τ_{NN} at 25.0 at% Al was higher than that at 23.0 at% Al by 30 MPa (Fig. 7), which may result in the highest R in Fe–25.0Al polycrystals.

The strain recovery of Fe–Al polycrystals was much lower than that of the single crystals (Figs. 3 and 4). For instance, the recoverable strain of Fe–23.0Al polycrystals was below 1.0%, much smaller than that of the single crystals. It is noted that Fe–23.0Al single crystals oriented for single slip exhibited a maximum recoverable strain.⁸⁾ In contrast, in a polycrystalline material, a sufficient number of independent slip systems are required to accommodate strain at a grain boundary. The activated slips in Fe–Al polycrystals interacted with each other resulting in the formation of sessile dislocations. Moreover, the grain boundaries suppressed the reversible motion of $1/4\langle 111 \rangle$ superpartials dragging the NNAPB. Thus, the formation of the sessile dislocations and the grain boundary effect led to the decrease in R in the polycrystals. In addition, in Fe–23.0Al polycrystals, the R rose with an increase in $\bar{d}$ as shown in Fig. 6. As the $\bar{d}$ of the polycrystals increased, the effect of the grain boundaries became small, which was suitable for the pseudoelasticity. However, the elongation of Fe–Al polycrystals decreased with increasing $\bar{d}$ due to the stress concentration at the grain boundaries. Therefore, an optimization of $\bar{d}$ is necessary for obtaining good balance between the pseudoelasticity and elongation.

The effect of crystallographic texture on the pseudoelasticity was discussed. In Fe–Al polycrystals, the recrystallization texture at 800°C varied from near-Cube to the γ fiber textures with increasing Al concentration as shown in Fig. 2. Below 23 at% Al where the near-Cube texture developed, the τ_{NN} was so small that reversible motion of $1/4\langle 111 \rangle$ superpartials hardly occurred irrespective of the texture (Fig. 7). Therefore, the texture transition was insignificant for the pseudoelastic properties of Fe–Al polycrystals. On the other hand, the pseudoelasticity of Fe–25.0Al polycrystals showed strong θ dependence (Fig. 5). The R and σ_y at $\theta=0^\circ$, 90° was much higher and lower than that at $\theta=45^\circ$, respectively. The dependence of R and σ_y on θ was found to correlate with Taylor factor (M) which could be obtained by the OIM software. Fe–25.0Al polycrystals warm-rolled and annealed at 800°C exhibited the γ fiber texture where $\langle 111 \rangle$ direction of the grains was oriented parallel to the ND. Although the formation of the γ fiber texture generally led to isotropic deformation in the polycrystalline sheets compared with the other texture components, plastic anisotropy depending on θ was reported in some bcc metals.⁹⁾ For instance, both the σ_y and M of the Al-killed steels having the γ fiber texture showed a maximum at around $\theta=45^\circ$. In the present study, the M of Fe–25.0Al polycrystals was also largest at $\theta=45^\circ$ among the three loading axes similar to the bcc metals (Fig. 8). Moreover, it is natural that large M at $\theta=45^\circ$ resulted in higher σ_y . The orientation dependence of the pseudoelasticity of the Fe–Al single crystals was systematically examined in our previous work.⁸⁾ In the single crystals, the R increased with increasing Schmid factor for primary

Fig. 8. Change in M with θ in Fe-25.0Al polycrystals recrystallized at 800°C for 1 h and then pulled at room temperature.

$\{101\}\langle 111\rangle$ slip. If the Schmid factor decreased, $1/4\langle 111\rangle$ superpartials tended to cross slip onto $\{211\}$ plane. Numerous dislocation dipoles which was immobile and suppressed the reversible motion of $1/4\langle 111\rangle$ superpartials were formed as a result of the cross slip. Thus, high Schmid factor for $\{101\}\langle 111\rangle$ primary slips was favorable for large strain recovery in Fe-Al single crystals. The large M at $\theta=45^\circ$ means the existence of $\{101\}\langle 111\rangle$ slip of which Schmid factor is small, which resulted in small R at the orientation. However, the variation in R with θ became unnoticeable after work hardening at and above $\epsilon_t=3.0\%$. It should also be noted that Young's modulus of bcc metals with the γ fiber texture showed a maximum at $\theta=45^\circ$.¹⁰⁾ High Young's modulus at $\theta=45^\circ$ led to an increase in plastic strain at constant ϵ_t , also resulting in the decrease in R . One of the possible methods to avoid the effects of grain boundary and crystallographic texture is to control an abnormal grain growth. It is widely known that strong $\{110\}\langle 001\rangle$ Goss texture of which orientation dispersion is within 3° can be obtained in Fe-3 mass%Si steels with the bcc structure by controlling the grain growth.¹¹⁾ If such strong texture is developed in Fe-Al polycrystals, the polycrystals can be regarded as the single crystal. This may lead to the improvement of the pseudoelasticity. Since Fe-Al alloys have the $D0_3$ structure based on the bcc structure, there is a possibility that the texture can be controlled by the microstructure control. Further work along this line is presently being undertaken.

5. Conclusions

The pseudoelastic behavior of Fe-Al polycrystals was examined focusing on Al concentration, the plastic anisotropy and the grain size. The following conclusions were reached.

(1) $D0_3$ -ordered Fe-Al polycrystals as well as the single crystals showed APB pseudoelasticity at room temperature. In particular, Fe-25.0Al polycrystals exhibited perfect strain recovery at $\epsilon_t=1.0\%$. On the other hand, the R became small as the deviation from 25.0 at% Al increased.

(2) The pseudoelasticity of Fe-25.0Al polycrystals was dependent on θ . The R at $\theta=45^\circ$ was lower than that at $\theta=0^\circ$ and 90° .

(3) The R increased with an increase in $\bar{d}$, presumably due to the suppression of the reversible motion of $1/4\langle 111\rangle$ superpartials by the grain boundaries.

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REFERENCES

- 1) K. Otsuka and C. M. Wayman: Shape Memory Materials. Cambridge University Press, Cambridge, (1998), 1.
- 2) L. P. Kubin, A. Fourdeux, J. Y. Guedou and J. Rieu: *Philos. Mag. A*, **46** (1982), 357.
- 3) H. Y. Yasuda, K. Nakano, T. Nakajima and Y. Umakoshi: *Acta Mater.*, **51** (2003), 5101.
- 4) H. Y. Yasuda, T. Nakajima, K. Nakano, K. Yamaoka, M. Ueda and Y. Umakoshi: *Acta Mater.*, **53** (2005), 5343.
- 5) F. J. Humphreys and M. Hatherly: Recrystallization and Related Annealing Phenomena, Elsevier, Oxford, (1995), 1.
- 6) I. L. Dillamore and H. Katoh: *Metall. Sci.*, **8** (1974), 73.
- 7) H. J. Leamy and F. X. Kayser, *Phys. Status Solidi*, **34** (1969), 765.
- 8) H. Y. Yasuda, K. Nakano, M. Ueda and Y. Umakoshi: *Mater. Sci. Forum*, **426-432** (2003), 1801.
- 9) H. J. Bunge and W. T. Roberts: *J. Appl. Cryst.*, **2** (1969), 116.
- 10) C. S. Barrett: Structure of Metals, 2nd ed., McGraw-Hill, New York, (1952), 534.
- 11) F. J. Humphreys and M. Hatherly: Recrystallization and Related Annealing Phenomena, Elsevier Science, Oxford, (1996), 407.