The Effects of Aging on the Degree of Order in Cu-Al-Ni Shape Memory Alloys

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The fractions of Al atoms at three kinds of crystallographic sites were determined in a Cu‐15.5Al‐4.0Ni (mass%) alloy before and after aging using powder X‐ray analysis in order to clarify the effect of aging on the martensite start temperature, \( M_s \). The ordering between first nearest neighbors (the B2 ordering) was almost finished even just after quenching, but the ordering between second nearest neighbors (the L21 ordering) was incomplete. The B2 type ordering occurs in advance of the L21 ordering in Cu‐Al‐Ni alloys as well as in Cu‐Al‐Zn alloys. The degree of L21 ordering increases from 0.699 to 0.844 during aging at 200°C for an hour. The increase in \( M_s \) during the aging is attributed to the L21 ordering in Cu‐Al‐Ni alloys. [doi:10.2320/matertrans.MB201511]

(Received September 7, 2015; Accepted October 27, 2015; Published December 11, 2015)

**Keywords:** aging, degree of order, copper-aluminum-nickel, shape memory alloy, powder X-ray diffraction, mechanical alloying

1. Introduction

Cu‐Al‐Ni alloys exhibit shape memory effects and super‐elasticity, which are both driven by thermoelastic martensitic transformation (TMT). In the case of Cu‐Al‐Ni alloys, TMT causes change from L21 to 2H structures.1,2) Cu‐Al‐Ni alloys as well as Cu‐Zn‐Al alloys have L21 ordered structure shown in Fig. 1. The structure of the Cu‐Al‐Ni alloy is often referred to as D03 structure but it should be regarded as L21 rather than D03 because Ni atoms prefer to occupy site III.3,5) The martensite start temperature, \( M_s \), varies depending on the compositions of the alloys, but it also varies depending on quenching rate or aging conditions. The effects of the aging on \( M_s \) are often discussed, associated with the changes in degree of order in parent phases. Rapacioli and Ahlers calculated that effects of the disordered pairs on the formation energy of both parent and martensite phases, and they concluded that disordered pairs between first nearest neighbors (FNN) cause the increase of \( M_s \) in Cu‐Zn alloys, and those between second nearest neighbors (SNN) cause the decrease of \( M_s \) in Cu‐Zn‐Al alloys.6) Nakamura et al. investigated that the relation between the ordering and \( M_s \) in Cu‐Al‐Ni alloys,7) by means of electrical resistivity measurement. They concluded that aging below 250°C promotes the L21 ordering in a Cu68Al28Ni4 alloy in advance of the precipitation of \( \gamma' \) phase.

Nakata et al. found that the X‐ray intensity ratio of the superlattice to fundamental lattice diffractions, \( I_s/I_f \), decreases with increasing the quenching rate.9) Moreover, they found that the decrease in \( I_s/I_f \) due to rapidly quenching was remarkable for the diffractions characteristic of the L21 structure such as \{111\} diffraction rather than for those of the B2 structure such as \{200\}. The former diffraction originates from the ordering between SNN, that is, between site I and site II in Fig. 1, whereas the latter originates from the ordering between FNN, that is, between site III and site I (or site II). The results of the X‐ray diffraction indicate that the B2 ordering is nearly finished even after rapid quenching but the L21 ordering is incomplete yet. The X‐ray measurement provided the direct evidence for the reason why the rapid quenched samples have lower \( M_s \) than the slow quenched samples.

Although the above X‐ray experiment shows the relativistic changes in degree of order between slow and rapid quenching samples, the degree of order itself was not determined. This is because the single crystal used in the above measurement did not have a proper size for X‐ray analysis. Small size samples about 0.1 mm in diameter are required for single crystal X‐ray diffractions to reduce the effects of absorption and extinction. In this meaning, powder samples are more suitable to investigate the effect of aging or quenching on degree of order.

Recently, we have succeeded in fabricating fine powder samples of Cu‐Al‐Ni alloys by mechanical alloying (MA) technique. Particle sizes of the powder samples obtained were about 1‐2 \( \mu \)m, which is enough small for powder X‐ray diffraction. In addition, the particles had a nearly spherical shape, so that the preferred orientation effects were expected to be small. In the present study, the effect of aging on the lattice constant and the crystal size were also investigated as well as degree of order, using powder X‐ray diffraction technique.

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2. Experimental Procedure

The powder samples with compositions of Cu-13.5 mass\% Al-4.0 mass\% Ni (Cu-26.8 at\% Al-3.7 at\% Ni) and Cu-15.5 mass\% Al-4.0 mass\% Ni (Cu-30.1 at\% Al-3.6 at\% Ni) were prepared from Cu (purity: 99.99\%, particle size: 150 µm), Al (99.99\%, 44 µm), and Ni (99.70\%, 74 µm) powders using MA technique. The container and grinding balls used in MA were made of austenitic stainless steel and the size of the ball is 11 mm in diameter. The total mass of the starting powder materials was 20 g, and the mass ratio of the powder to the balls was about 1 to 21. Methyl alcohol was used as a process-control agent in MA processing. The MA processing to yield powders using MA technique. The container and grinding balls used in MA were made of austenitic stainless steel and the size of the ball is 11 mm in diameter. The total mass of the starting powder materials was 20 g, and the mass ratio of the powder to the balls was about 1 to 21. Methyl alcohol was used as a process-control agent in MA processing. The MA processing was performed for 70 hours in the cylindrical container vibrated at 12.0 Hz. The container was filled with argon gas (99.9\%) and cooled by water throughout the MA processing. After the MA processing, the container was first evacuated for several minutes to remove the methyl alcohol, and then filled with argon gas (99.9\%). The MA powders were collected later than 24 hours to avoid explosive oxidation. The particle sizes of the MA powders were determined to be about 1 to 2 µm by scanning electron microscopy (SEM). The alloying process leads to a considerable reduction in the particle size of the starting materials. Each MA powder was annealed at 1000°C for 1 hour for homogenization in an evacuated quartz tube and then quenched into iced water.

X-ray diffraction measurements were carried out at room temperature on an automatic X-ray powder diffractometer with a Bragg-Brentano goniometer and a secondary graphite monochromator (Rigaku RINT2000) using a copper X-ray tube. The X-ray samples were scanned in the range of 10° to 160° in 2θ with a constant step of 0.02° in 2θ and with a fixed counting time of 2 s.

3. Analysis of Degree of Order in a Cu-Al-Ni Alloy by Powder X-ray Diffractions

The Bragg peak widths changed irregularly with the diffraction angles. Thus, each peak was fitted separately with a pseudo-Voigt function, $\varphi$, expressed by the following equation,\(^9\)

$$
\varphi(\Delta 2\theta) = \frac{2}{\pi W} \left[ 1 + d \left( \frac{\Delta 2\theta}{W} \right)^2 \right]^{-1} + (1 - d) \frac{2 \sqrt{n/2 \ln 2}}{\pi WN} \exp \left[ -4 \ln 2 \left( \frac{\Delta 2\theta}{W} \right)^2 \right],
$$

where $\varphi$ is expressed as a function of $2\Delta\theta$, which is a deviation from the Bragg peak position, $2\theta_0$, that is, $2\Delta\theta = 2\theta - 2\theta_0$. In eq. (1), the first and second terms in the right side are the Lorentzian and the Gaussian, respectively, with the peak width of $W$, which are mixed in the ratio of $n:1-n$. Using the above function, $\varphi(\Delta 2\theta)$, the diffracted intensity, $I_{\varphi}$, at any angle, $2\theta$, can be approximated by a superposition of $K_1$ and $K_2$ diffracted intensities and background as follows.

$$
I_{\varphi}(2\theta) = I_{\varphi}(\Delta 2\theta) + \frac{1}{2} I_{\varphi}(\Delta 2\theta - 2(2\theta_0)) + C_1 \cdot 2\theta + B_0,
$$

where $\delta$ gives the separation angle of $K_1$ (0.154438 nm) and $K_2$ (0.154060 nm). In the right-hand side of the above equation, the first and second terms are X-ray intensities of $K_1$ and $K_2$, of which ratio is fixed to 0.5, and the third one is a linearly sloping background intensity. The fitting of the function given in eq. (2) to the experimental data was performed using Marquart method, one of non-linear least squares fitting methods. The integrated peak intensity, $I$, obtained from the curve fitting using eq. (2) is expressed in the following formula,

$$
I = A |F_{hkl}|^2 L_p \exp \left[ -2B \left( \frac{\sin \theta}{\lambda} \right)^2 \right],
$$

where $A$, $F_{hkl}$, $p$, $L_p$, $B$, and $\lambda$ are an experimental constant proportional to the incident X-ray intensity, the structure factor of a $(h \ k \ l)$ plane, the multiplicity of a $(h \ k \ l)$ plane and the Lorentz-polarization factor, the Debye–Waller factor and the wavelength of X-ray. In eq. (3), the Lorentz-polarization factor is expressed by the following formula,

$$
L_p = \frac{1 + \cos^2 2\theta_m \cos^2 2\theta}{\sin^2 \theta \sin^2 \theta},
$$

where $\theta_m$ and $\theta$ are the Bragg angles of the monochromator and the samples, respectively. The structure factor, $F_{hkl}$, for the $L_2_1$ structure is not zero if $h$, $k$, and $l$ are all odd or even, and can be classified into the following three types,

- $F_{hkl} = 4(f_1 + f_{II} - 2f_{III})$ for $h + k + l = 4n + 2$,
- $F_{L2_1} = 4(f_1 - f_{II})$ for $h + k + l = 4n + 1$,
- $F_I = 4(f_1 + f_{II} + 2f_{III})$ for $h + k + l = 4n$.

In the above equations, $f_1$, $f_{II}$, and $f_{III}$ are the atomic form factors for sites I, II, and III in Fig. 1, respectively; each of them is averaged over atoms occupying each site. Since $F_{B2}$ expresses the difference in the form factor between the corner sites (sites I and II) and the body center sites (site III), this type diffraction depends on the B2 ordering. On the other hand, $F_{L2_1}$ expresses the difference in the form factor between sites I and III, This type diffraction depends on the $L_2_1$ ordering.

The values of $f_1$, $f_{II}$, and $f_{III}$ are determined by the compositions of sites I, II and III, respectively, which are shown in Table 1. The values of $g_1$ and $g_2$ in Table 1 are the fraction of Al atoms at sites I and II, respectively; Ni atoms are assumed to occupy only site III.\(^3\) Using these values, the structure factors given in eq. (5) are rewritten as follows:

- $F_{B2} = 2[2c_{Al} + (g_1 + g_2)(f_{Al} - f_{Cu}) + 4c_{Ni}(f_{Cu} - f_{Ni})]$ for $h + k + l = 4n \pm 2$,
- $F_{L2_1} = (g_1 - g_2)(f_{Al} - f_{Cu})$ for $h + k + l = 4n + 1$,
- $F_I = 4[c_{Al}f_{Al} + c_{Ni}f_{Ni} + (1 - c_{Al} - c_{Ni})f_{Cu}]$ for $h + k + l = 4n$.

Since $F_I$ is independent of $g_1$ and $g_2$, the values of $A$ and $B$ in eq. (2) can be determined using the diffraction intensities satisfying the condition of $h + k + l = 4n$. Once the values of $A$ and $B$ were determined, the X-ray intensity, $I$, in eq. (3) can be expressed by only $g_1$ and $g_2$.

Here, we define degrees of the $B2$ ordering and the $L_2_1$ ordering as the following equations,
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Table 1  Fraction of each element at sites I to III. The values of \( g_1 \) and \( g_2 \) are the fraction of Al atoms at sites I and II, respectively, and \( c_{Al} \) and \( c_{Ni} \) are the mole fractions of Al and Ni atoms in a Cu-Al-Ni alloy. In the case of Cu-15.5Al-4.0Ni (mass%), \( c_{Al} = 0.301 \) and \( c_{Ni} = 0.036 \). The last row shows the averaged value of each element over sites I to III.

<table>
<thead>
<tr>
<th>Site</th>
<th>Al</th>
<th>Cu</th>
<th>Ni</th>
</tr>
</thead>
<tbody>
<tr>
<td>I</td>
<td>( g_1 )</td>
<td>( 1 - g_1 )</td>
<td>0</td>
</tr>
<tr>
<td>II</td>
<td>( g_2 )</td>
<td>( 1 - g_2 )</td>
<td>0</td>
</tr>
<tr>
<td>III</td>
<td>( 2c_{Al} - g_1/2 - g_1/2 )</td>
<td>( 1 - 2c_{Ni} - 2c_{Al} + g_1/2 + g_1/2 )</td>
<td>( 2c_{Ni} )</td>
</tr>
<tr>
<td>1/4 + II/4 + III/2</td>
<td>( c_{Al} )</td>
<td>1 - ( c_{Al} - c_{Ni} )</td>
<td>( c_{Ni} )</td>
</tr>
</tbody>
</table>

Fig. 2  X-ray diffraction patterns for the as-quenched Cu-13.5Al-4.0Ni alloy (a) and the aged one at 200°C for an hour (b). The underline indicates the index of 2H structure.

\[
S_{B2} = \frac{g_1 + g_2 - 2c_{Al}}{2c_{Al}}, \\
S_{L21} = g_1 - g_2. 
\]

Following this definition, \( S_{B2} \) and \( S_{L21} \) vary in the range of 0 to 1. For example, \( S_{B2} = S_{L21} = 0 \) when Al atoms occupy randomly sites I to III. On the other hand, \( S_{B2} = 1 \) when no Al atoms occupy site III, that is, \( 2c_{Al} - g_1/2 - g_1/2 = 0 \). When \( g_1 = 1 \) and \( g_2 = 0 \), \( S_{L21} = 1 \), but \( S_{B2} \neq 1 \). In general, it is impossible that \( S_{B2} = S_{L21} = 1 \) for \( c_{Al} \neq 0.25 \).

4. Results

Figure 2 shows the X-ray diffraction patterns for the as-quenched Cu-13.5Al-4.0Ni alloy and the aged one at 200°C for an hour. The diffraction peaks recognized in both figures can be all indexed in terms of the L21 structure of the parent phase and the 2H structure of the martensite phase.\(^1\),\(^2\) This shows that powder samples prepared by the MA technique have the same crystal structures as bulk samples prepared by a conventional melting method. In Fig. 2(a), the main peaks are all identified as the diffraction of the parent phase with the L21 structure, except from a couple of minor peaks. Thus, the martensite start temperature, \( M_s \), of the as-quenched sample can be estimated to be slightly above room temperature. This shows that the sample used in the present study have the composition nearly equal to the mixing ratio of the starting materials, because \( M_s \) of a bulk sample with the designing composition is around 60°C.\(^10\),\(^11\)

In contrast to the as-quenched sample, most of diffraction peaks for the aged sample is due to the 2H martensite structure as shown in Fig. 2(b). The increase in the amount of martensite due to the aging can be attributed to the increase in \( M_s \) although \( M_a \) of the MA powder samples was not measured directly. The aging brings about the increase in \( M_s \) for the MA powder samples as well as for bulk samples.

Figure 3(a) and (b) show the X-ray patterns for an as-quenched Cu-15.5Al-4.0Ni powder sample and for the aged one at 200°C for an hour, respectively. Since this sample contains 2 mass% more Al than the sample shown in Fig. 2, the martensite phase does not appear even after aging. Thus, this sample can be used to investigate the effects of aging on the B2 and L21 ordering in the parent phase. In Fig. 3, the intensity ratio of the \{1 1 1\} and \{2 0 0\} diffraction peaks increases during aging. On the basis of eq. (5), this result shows that the L21 ordering proceeds during the aging. On the other hand, the \{2 0 0\} diffraction, which originates from the B2 ordering, does not vary so remarkably as the \{1 1 1\} diffraction. This means that degree of the B2 ordering is nearly perfect even in the as-quenched sample. However, degree of the L21 ordering is incomplete just after quenching and it increases during aging.

4. In order to investigate the quantitative change of the B2 and L21 orderings, first, the integral intensities of the diffraction peaks were determined by the curve fitting. As shown in Fig. 4, pseudo-Voigt functions fit into all diffraction patterns ranging from a broad peak such as \{1 1 1\} to a narrow peak such as \{2 0 0\}. The parameters obtained from curve fitting are shown in Table 2.

Next, the values of \( A \) and \( B \) in eq. (3) were determined using the values of \( I \) for fundamental lattice diffractions, of
which $F_{hkl}$ are independent of $g_1$ and $g_2$. Figure 5 shows the plot of $\log(I/pL_{p}[F_{hkl}]^2)$ against $(\sin \theta/\lambda)^2$ for the as-quenched and aged samples. Since the data points plotted in Fig. 5 lie on a straight line, the values of $A$ and $B$ in eq. (3) can be determined with great accuracy by the least square fitting. As seen in Fig. 5, the as-quenched sample has almost the same values of $A$ and $B$ as the aged one. In Fig. 5, little deviation from straight line shows that the effects of preferred orientation on the peak intensities are negligible small.

Once the values of $A$ and $B$ were determined, the X-ray intensity $I$ in eq. (3) can be expressed by only $g_1$ and $g_2$. Since the peak intensities of the $\{311\}$ and $\{222\}$ diffractions are very weak, $g_1$ and $g_2$ were determined by only $\{111\}$ and $\{200\}$ diffractions. Table 3 shows the fraction of Al at sites I to III obtained from as-quenched and aged samples. In addition, the degrees of order, $S_{B2}$ and $S_{L21}$, are also determined by eq. (7). The change in $S_{B2}$ due to the aging is little but that in $S_{L21}$ is considerably large. Although $S_{B2}$ is not perfect before aging, it is nearly constant during aging. On the other hand, the $L2_1$ ordering increases more than 15% during aging.

5. Discussion

5.1 Degree of order

As shown in Table 3, $S_{B2}$ is nearly constant during aging. On the other hand, $S_{L21}$ increases from 0.699 to 0.844. These results show that the ordering between FNN (the $B2$ ordering) was almost finished even just after quenching, but the $L2_1$ ordering is incomplete. Therefore, the $B2$ type ordering occurs in advance of the $L2_1$ ordering. This fact can
be understood from the difference in the ordering temperatures. According to the recent study, the disordered $\beta$ phase first changes to B2 phase around 810 K and then B2 phase changes to L2$_1$ phase around 740 K in Cu-26.48Al-3.65Ni (mass%).

According to Rapacioli and Ahlers, the B2 ordering in Cu-Zn-Al alloys stabilizes the parent phase rather than the martensite phase, resulting in decrease in $M_s$. On the other hand, the L2$_1$ ordering stabilizes the martensite phase rather than the parent phase, resulting in increase in $M_s$. The phase stability due to the L2$_1$ ordering originates from the fact that a part of the SNN pairs change to the FNN pairs in the martensite phase. Since the increasing disordered pairs between SNN in the L2$_1$ structure bring about the decrease in the Cu(Zn)-Al bonding pairs in the martensite phase, the martensite phase becomes more unstable than the parent phase. On the contrary, the increase in the L2$_1$ ordering makes the martensite phase more stable than the parent phase. This situation is also right in Cu-Al-Ni alloys because of similarity between 2H and 18R. It is concluded that the increase in $M_s$ during aging is caused by the increase in $S_{L21}$. The present study gives the direct evidence for the relation between $S_{L21}$ and $M_s$.

5.2 Lattice constant

The lattice constants obtained from Bragg angles, $\theta_B$, in Table 2 are dependent on diffraction indices because the values of $\theta_B$ contain some systematic errors. In order to eliminate the errors, the lattice constants are plotted against the Nelson-Riley function. The accurate lattice constant can be determined by the extrapolation to $\theta = 90^\circ$. The lattice constants obtained from $\theta_B$ in Table 2 are plotted in Fig. 6. The data points make straight lines for both as-quenched and aged samples; the values extrapolated are 0.58358 $\pm$ 0.00008 nm for the as-quenched one and 0.58333 $\pm$ 0.00006 nm for the aged one. The lattice constant decreased about 0.04% during aging. The decrease in the lattice constant is slight but significant. The similar behavior was also observed in Cu$_3$Au. Lattice constants obtained for disordered and ordered Cu$_3$Au alloys ($S = 0.94$, where $S$ is degree of order) are 3.7490 $\pm$ 0.0003 and 3.7426 $\pm$ 0.0003, respectively, at room temperatures. Owing to the ordering, the lattice constant of Cu$_3$Au decreases about 0.17%. The slight decrease in lattice constant of the Cu-Al-Ni alloy is reasonable because the change in the L2$_1$ ordering is only about 15%.

Annealing out of quenched vacancies is often regarded as a factor of changes in $M_s$. It is possible that the aging may decrease quenched vacancies. However, decrease of vacancies is expected to increase the lattice constant. Since the lattice constant decrease in the present study, vacancies are considered not to change during aging.

5.3 Particle size

In general, peak widths of X-ray diffraction peaks provide us the information for crystallite sizes of X-ray samples. However, the peak widths are also affected by local strain in the crystallite. According to Williamson and Hall, the peak width, $W_{hkl}$, at the peak position, $2\theta_{hkl}$, can be expressed by the following formula,

$$W_{hkl} \frac{\cos \theta_{hkl}}{\lambda} = 2\varepsilon \frac{\sin \theta_{hkl}}{\lambda} + \frac{K}{D},$$

where $\varepsilon$ and $D$ are the local strain and the crystallite size. In eq. (8), $K$ is a constant depending on the crystallite habit etc., which is usually taken as 0.9 to 1. In Fig. 7, the values of $W \cos \theta/\lambda$ are plotted against $\sin \theta/\lambda$, using $W$ and $\theta_B$ shown in Table 2. The data points plotted in Fig. 7 are expected to be on a straight line as seen in eq. (8). Thus, the two straight lines are drawn between the data points of $\{n 0 0\}$ type diffractions and between those of $\{n n 0\}$ type diffractions. According to eq. (8), the crystallite size, $D$, can be obtained from y-intercept of the extrapolated line. The values of $D$ estimated from $\{n 0 0\}$ and $\{n n 0\}$ are 43 and 83 nm, respectively. These values indicate thicknesses of crystallites in $(1 0 0)$ and $(1 1 0)$ directions. It is noted that these values are rough approximations because effects of X-ray diffraction instrument on broadening is not taking into account in the above analysis.

The $(1 1 1)$ diffraction as well as the other diffractions exhibits little change in its peak width during aging. This means that the L2$_1$ ordering domains hardly grow during aging in spite of the increase in $S_{L21}$. This may be because the L2$_1$ ordered domains have already grown to the boundaries of the crystallites before aging. Assuming that the $\{n n n\}$ type diffractions should be on a straight line with the same slope.

Fig. 6 The lattice constants obtained from the data shown in Table 2 are plotted against the Nelson-Riley function.

Fig. 7 $W \cos \theta/\lambda$ are plotted against $\sin \theta/\lambda$, using $W$ and $\theta_B$ shown in Table 2.
as the \( \{n 0 0\} \) and \( \{n n 0\} \) type diffractions, the thickness of the crystallites in the \( \{1 1 1\} \) directions is roughly estimated to be about 20 nm.

The direction dependence of the crystallite thickness can be understood from the difference in atomic sites composing the corresponding atomic planes. The \( \{2 0 0\} \) planes perpendicular to the \( \{1 0 0\} \) direction consist of two kinds of atomic planes. One is composed of sites I and II, and the other is composed of only site III. In a similar way, the \( \{1 1 1\} \) planes consist of three kinds of atomic planes. Each plane is composed of only one of site I, II and III. Thus, the planes perpendicular to \( \{1 0 0\} \) and \( \{1 1 1\} \) directions consist of more than two kinds of atomic planes. Such atomic planes are generally unstable and they are usually not chosen as facets of crystallites. On the other hand, both \( \{2 2 0\} \) and \( \{4 2 2\} \) planes consist of only one kind of atomic plane which is composed of all of sites I to III. In such a case, atomic planes are stable. Thus, the facets of crystallites are considered to be \( \{2 2 0\} \) and \( \{4 2 2\} \) planes. The crystallites grow easily in the \( \{1 1 0\} \) and \( \{2 1 1\} \) directions. The broad peaks observed in the present work are concerned not with ordered domains but with the growth mechanism of the crystallites fabricated by MA technique.

6. Conclusion

The ordering behavior is investigated using the fine powder sample fabricated by MA technique. As the result, the following conclusions are obtained,

(1) The fine powder samples show the increase in \( M_s \) due to aging as well as bulk samples fabricated by conventional melting methods.

(2) The B2 ordering have already finished just after quenching, but the L21 ordering is incomplete. It proceeds remarkable during aging at 473 K for 1 hour. Accordingly, the lattice constant of the parent phase decreases.

(3) The increase in \( M_s \) during the aging can be attributed to the increase in degree of the L21 ordering, which stabilizes the martensite phase with the 2H type structure.

(4) The crystallites of the powder samples have different thicknesses, depending on the crystal directions. This is concerned with the mechanism of crystal growth.

REFERENCES