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Effect of Ultrasound Treatment on Age-Hardening Response in an Al-Mg-Si Alloy

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An attempt to improve age-hardening response in an Al-Mg-Si alloy has been made in the temperature range of room temperature (RT) to 443K by exploiting ultrasound which penetrates silicone oil in a bath with a frequency of ~1MHz. The application of ultrasound treatment has little effect on natural-aging behavior of the alloy sheet, but increases the age-hardenability by ~5Hv at 373K and by ~20Hv at 443K. Such an increase in mechanical strength is suitable for the fabrication of automobile body panels, and therefore the mechanism is expected to be clarified not only from an academic point of view but also from an industrial point of view. In this work, two possible effects of ultrasound; i.e. vibrational force by a liquid medium of silicone oil and shock wave by the collapse of cavitation bubbles, have been quantified to clarify the mechanism by which the concentration of vacancies, diffusion of atoms and/or nucleation and growth of nano-scale precipitates (nanoclusters) might be influenced.

Keywords: *Al-Mg-Si alloy ,ultrasound, age-hardening, microstructure, nanocluster*

1. Introduction

Heat treatable Al-Mg-Si alloys are used for automobile body panels because of their good formability, corrosion resistance, surface quality and precipitation strengthening after paint-bake treatment at ~443K. It is well known that the alloys containing excess of Si over Al-Mg_2Si quasi-binary composition; e.g. AA6022 alloy, exhibit a pronounced age-hardenability due to the formation of the β" phase and/or its precursors; e.g. nanoclusters called Cluster(1) and Cluster(2) [1]. However, more increased age-hardening response is anticipated to meet industrial demands for high strength and good dent resistance comparable to those of the conventional steel-made body panels.

Ultrasound is a clean and safe energy, and therefore a wide range of applications have been developed in the modern industries. The authors reported for the first time that the application of ultrasound during heat treatments of Al-4%Cu and Al-4%Cu-0.5%Mg alloys complexly changes their age-hardenability [2]. It can be hypothesized that ultrasound assists in atomic vibration in the alloys, promotes diffusion of atoms and therefore results in the accelerated precipitation from super saturated solid solutions. However, it is also feasible to consider that the more excited atomic vibration inversely inhibits the formation of nanoclusters, leading to suppressed age-hardening response.

In this work, the effect of ultrasound on age-hardening response of AA6022 alloy has been investigated to confirm which of improvement or degradation in age-hardenability is achieved by the newly developed ultrasound treatment. The mechanism of such an effect of ultrasound treatment was also quantitatively discussed from the viewpoints of vibrational force by a liquid medium of silicone oil and shock wave by the collapse of cavitation bubbles.

2. Experimental

The chemical composition of the alloy utilized in this work is listed in Table 1. The alloy ingot was homogenized, hot- and cold-rolled through the standard processes to the final thickness of 1.0mm. The sheet specimens were solution heat-treated at 823K for 60s, and then water quenched. Aging treatment was carried out at RT, 373 and 443K in a silicone oil bath equipped with an ultrasound transducer (Fig.1). The specimen with dimensions of 15×15×1 mm^3 was fixed within silicone oil in contact with a thermocouple to keep a constant temperature, and vibrated by ultrasound penetrated from silicone oil. The frequency and amplitude of the vibration at the upper side of the specimen were 960kHz and 4.4nm at 443K, as measured by a laser Doppler vibration meter.

Hardness test was conducted by a Vickers hardness tester with 98N load and dwell time of 30s to compare age-hardening response of the specimens with and without ultrasound treatment. Increment in electrical resistivity during aging $\Delta\rho$,

$$\Delta\rho = 17.24 / \sigma \times 100, \quad (1)$$

was also compared through electrical conductivity σ (in IACS%) measured by an eddy current tester.

Table 1 Chemical composition of the alloy utilized in this work [in mass%].

Mg	Si	Fe	Mn	Ti	Cu	Al
0.53	1.05	0.15	0.07	0.02	0.01	Bal.

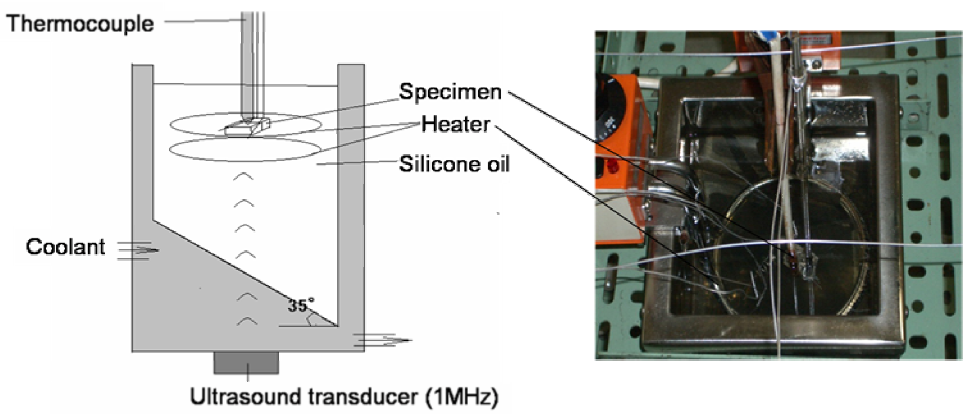

Fig.1 Schematic illustration and outlook of the ultrasound bath utilized in this work.

3. Results

3.1 Age-hardening response

Isothermal aging curves of hardness at RT, 373 and 443K are shown in Fig.2 for the specimens with and without ultrasound treatment. The application of ultrasound treatment has little effect on natural-aging (Fig.2(a)), but increases the age-hardenability at 373K by ~5Hv (Fig.2(b)). At 443K, furthermore, age-hardening response is significantly improved by ~20Hv, suggesting that ultrasound treatment is useful for the fabrication of aluminum-made body panels with higher strength and better dent resistance.

3.2 Electrical resistivity change

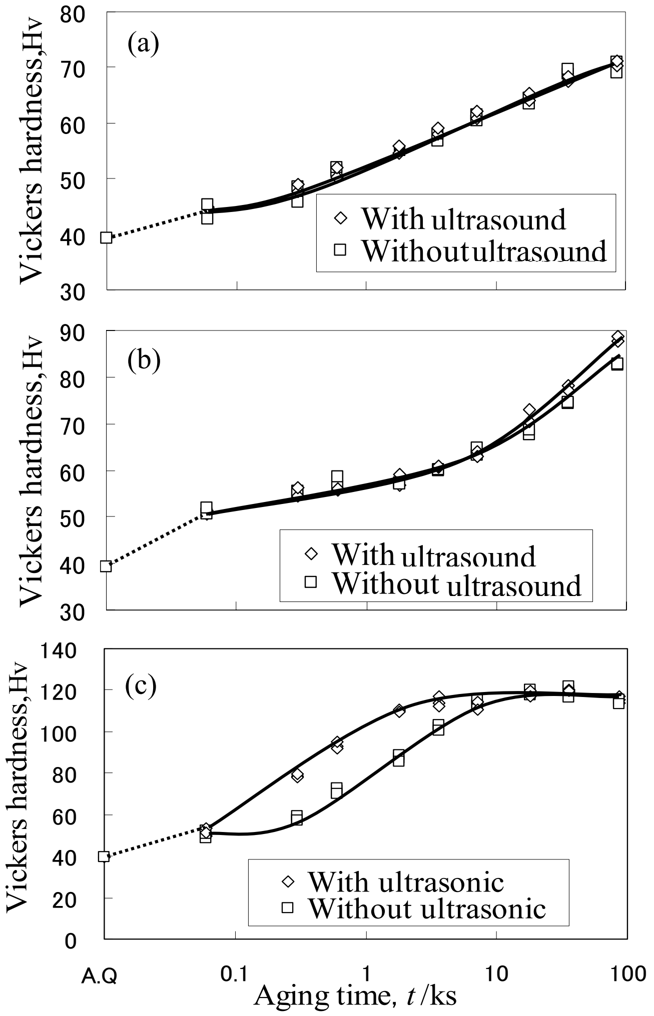

Fig.2 Age-hardening curves of the specimens with and without ultrasound treatment at (a)RT, (b)373K and (c)443K.

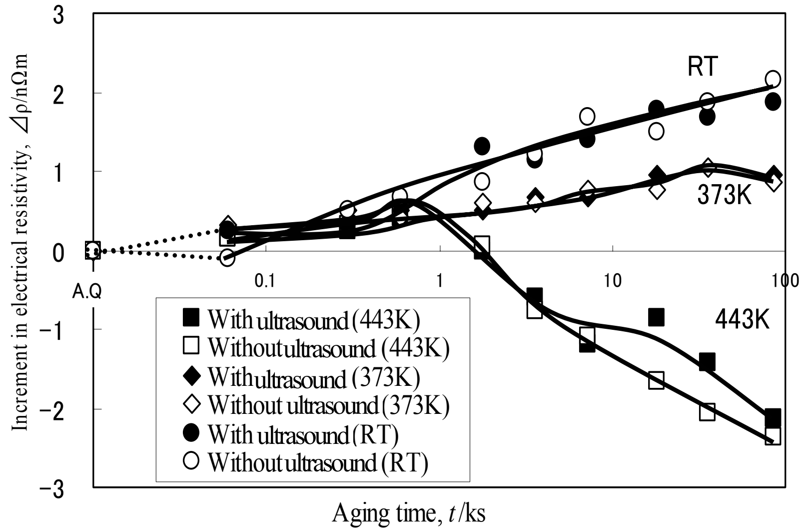

Fig.3 Increment in electrical resistivity of the specimens with and without ultrasound treatment at RT, 373 and 443K.

Fig.3 shows the increment in electrical resistivity Δρ of the specimens with and without ultrasound treatment at RT, 373 and 443K. The specimen with ultrasound treatment exhibits sluggish decrease in Δρ at 443K compared to the specimen without ultrasound treatment (see at aging times of >1.2ks). In general, it is believed that Δρ arises from the superimposition of the increases in both size and number density of precipitates, and the decrease in residual solute concentrations in the matrix [3]. Therefore, the sluggish decrease in Δρ is likely to indicate that refined precipitates distribute more densely in the specimen with ultrasound treatment. This interpretation well agrees with the fact that greater increases in hardness are observed with ultrasound treatment in Fig.2(b),(c). Note that microstructural observation by transmission electron microscopy (TEM) is quite difficult to capture any differences in morphology and distribution of such refined precipitates.

4. Discussion

Based on results in Figs.2 and 3, it can be concluded that ultrasound treatment has positive effect on age-hardening response of the Al-Mg-Si alloy at 373 and 443K. In this chapter, two possible effects of ultrasound; i.e. vibrational force by a liquid medium of silicone oil and shock wave by the collapse of cavitation bubbles, have been quantified to clarify the mechanism of such an effect of ultrasound treatment.

4.1 Vibrational force by a liquid medium of silicone oil

As the frequency f_0 and amplitude A_0 of vibration at the upper side of the specimen were measured to be 960kHz and 4.4nm at 443K, the maximum velocity $v_{\max}$ of silicone oil can be estimated through

$$v_{\max}=(2\pi f_0)\,A_0 = 2.65\times 10^{-2}\ \text{[m/s]}. \tag{2}$$

In general, the sound pressure P_0 and velocity v_0 at the upper side of the specimen are related to the sound pressure P_i and velocity v_i at the lower side of the specimen;

$$\begin{pmatrix} p_i \\ v_i \end{pmatrix} = \begin{pmatrix} \cos k_a t & -jp_a c_a \sin k_a t \\ -\dfrac{j}{\rho_a c_a}\sin k_a t & \cos k_a t \end{pmatrix} \begin{pmatrix} p_0 \\ v_0 \end{pmatrix}, \tag{3}$$

where k_a=$2\pi f_0/C_a$, C_a, ρ_a and t are wave number (1185[m^{-1}]), speed of sound (5092[m/s]) in the specimen at 443K, the density (2700[kg/m^3]) and thickness (1×10^{-3}[m]) of the specimen, respectively. P_i is therefore expressed as

$$p_i = jv_{\max}\left(\rho_s c_s \cos k_a t + j\rho_a c_a \sin k_a t\right), \tag{4}$$

because ultrasound wave penetrated from the specimen is no longer reflected in silicone oil. Here, C_s and ρ_s are speed of sound in silicone oil (663[m/s]) and the density of silicone oil (841[kg/m^3]) at 443K. It was found from such estimation that the specimen is subjected to stress $|P_i|$

$$|p_i| = v_{\max}\sqrt{\left(\rho_s{}^2 c_s{}^2 \cos^2(k_a t) + \rho_a{}^2 c_a{}^2 \sin^2(k_a t)\right)} = 338\ \text{[kPa]}, \tag{5}$$

and elastically deformed by strain of 4.8×10^{-6} because Young's modulus of the Al-Mg-Si alloy is around 70GPa at 443K. Note that such a stress is repeatedly applied to the specimen with a frequency of 960kHz.

4.2 Shock wave by the collapse of cavitation bubbles

Ultrasound achieves its chemical and/or mechanical effects by generating bubbles within a liquid medium, a process called cavitation. The vibrational energy by ultrasound causes the molecules in the liquid to be alternately compressed and stretched to produce bubbles. These bubbles are subjected to vibrational stresses and eventually collapse. Although there are no visible cavitation bubbles in the utilized ultrasound bath (Fig.1), the maximum pressure of shock wave P_{max} by the collapse of cavitation bubbles could be estimated through

$$P_{max} = P_g \left\{ \frac{P_e(\gamma - 1)}{P_g} \right\}^{\gamma(\gamma-1)}, \qquad (6)$$

under the assumption that a bubble is formed with the critical radius r_e determined by cativation threshold; i.e. 0.046μm, and the bubble grows up to the radius r of 0.1μm before the collapse [4]. P_e and P_g are effective and internal pressures of the bubble (in this work P_e was regarded as $|P_i|$, whereas P_g was calculated by $P_g = (P_1 + 2\sigma/r_e - P_v)\ (r_e/r)^{3K}$ where P_1 is hydrostatic pressure of 165Pa, σ is surface tension of 20.3mN, P_v is vapor pressure of 133Pa and K=1 because of the isothermal change) and γ is the ratio of specific heats (if the bubble is assumed to be filled with O_2, $\gamma = C_p/C_v = 1.40$), respectively. It was found from such estimation that the specimen is subjected to a comparable stress of P_{max} =111kPa to that by the above mentioned vibrational force, but probably with a much lower frequency.

4.3 Mechanism of accerelated precipitation by ultrasound treatment

It is obvious that age-hardening response of the Al-Mg-Si alloy is attributed to precipitation phenomena involving both of diffusion of Mg and Si atoms by the aid of quenched-in excess vacancies, and nucleation and growth of nano-scale precipitates. The application of ultrasound treatment was found to improve the age-hardenability of the alloy (Figs.2 and 3), and therefore it is plausible to consider that ultrasound influences either or both of them. The quantitative estimation of the effects of ultrasound treatment suggested that both of vibrational force by silicone oil and shock wave by the collapse of cavitation bubbles exert small stresses on the specimen in the order of 10^2kPa. These stresses are macroscopically within those generating only elastic deformation, but atomistic behaviors occurring in the alloy; e.g. formation of vacancies, diffusion of atoms, nucleation and growth of nanoclusters, might be influenced. Further investigation on the effects of ultrasound will be needed to clarify the mechanism of ultrasound treatment.

5. Conclusions

The effect of ultrasound treatment on the age-hardenablity of AA6022 alloy has been investigated. Although there was no significant difference in age-hardening response at RT, the increase in hardness during aging at 373 and 443K was found to be improved by the application of ultrasound treatment. This suggests that the newly developed ultrasound treatment is useful for the fabrication of aluminum-made body panels with higher strength and better dent resistance. The quantitative estimation of the effects of ultrasound treatment suggested that both of vibrational force by silicone oil and shock wave by the collapse of cavitation bubbles exert small stresses on the specimen in the order of 10^2kPa. Although these stresses are macroscopically within those generating only elastic deformation, atomistic behaviors occurring in the alloy; e.g. formation of vacancies, diffusion of atoms, nucleation and growth of nanoclusters, might be influenced. Further investigation on the effects of ultrasound treatment is in progress.

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