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Process-controlled suppression of natural aging in an Al–Mg–Si alloy

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In this study natural aging of an Al–Mg–Si alloy was investigated using various quenching processes. Atom probe tomography and electrical resistivity measurements reveal that solute clustering during natural aging can be suppressed by interrupting quenching for 120 s at 160 °C. This phenomenon is elucidated by simulating the excess vacancy annihilation. Reduced frozen-in excess vacancy concentration after the interrupted quenching can explain this experimentally observed suppression of natural aging.

Keywords: Aluminum alloys; Aging; Precipitation; Vacancies; Phase transformation kinetics

Al–Mg–Si alloys are the most widely used age-hardenable aluminum alloys [1,2], and many researchers have contributed to an understanding of aging phenomena in these alloys. Nevertheless, several important questions such as the characteristics of the major hardening phase [3] or the nature of processes that occur during storage at room temperature after quenching (natural aging) are still subjects of debate [4]. Natural aging, in particular, has emerged as a research field in recent years, because it can significantly affect industrial hardening strategies and consequently the resulting properties [2,5]. From an academic point of view surprisingly little is known about the early stages of solute clustering in this important class of aluminum alloys. Banhart et al. [6] reported that natural aging of Al–Mg–Si alloys is complex and takes place in up to five distinct, but still not understood, stages. A visualization of early-stage clusters is difficult because transmission electron microscopy and various scattering techniques do not produce distinct contrast [4,7]. Here, atom probe tomography (APT) is the most suitable technique for visualizing and measuring clusters in Al–Mg–Si alloys.

Frozen-in excess vacancies play an important role in the aging of Al–Mg–Si alloys, but are also known to be of crucial importance in other aluminum alloys, e.g. Al–Cu–Mg [8–10]. Rapid kinetics of natural aging in aluminum alloys was first explained by a simple expression of low-temperature diffusion after quenching, i.e. $D \propto \exp(E_m/RT) \times \exp(E_f/RT_s)$, where E_m is the migration barrier for diffusion, E_f is the formation energy of vacancies, R is the gas constant, T is the actual temperature and T_s is the temperature of the solution treatment prior to quenching [11]. This expression predicts that kinetics will scale with the equilibrium vacancy concentration at the solution-treatment temperatures. Although recent models [12] are more complex and the frozen-in excess vacancy concentration is not expected to be that high [13,14], the concentration of vacancies is important for the kinetics of all diffusional processes that occur at low temperatures, i.e. natural aging. The quenching process is expected to significantly affect the freeze-in of excess vacancies. As early as in the 1960s it was shown that Al–Cu alloys can be influenced in their clustering kinetics by a short interruption of quenching [11], and the effect was attributed to annihilation of excess vacancies. Despite intense activity in the field of

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Al–Mg–Si alloys in the last few years, however, no corresponding study has yet been published for this class of alloys. In the following we show that a modified quenching procedure can indeed be used to alter solute clustering in a lean Al–Mg–Si alloy during natural aging. This may be useful in controlling and understanding the effect of solute clustering on hardening processes in industrial Al–Mg–Si alloys.

The alloy studied (Al0.32Si0.32 Mg; all values given in wt.%) was prepared from pure Al (99.99%), Si (99.999%) and Mg (99.9%) using an inductive melting furnace. Argon gas purging was applied to reduce the hydrogen content before the alloy was cast to a slab. After cutting and homogenization (24 h at 550 °C), this slab was hot rolled at 550 °C. Finally, 3 mm rods were machined to produce wires of 1 mm in diameter by drawing. The wires were then heat-treated at 570 °C for 22 h under Ar atmosphere to generate a controlled bamboo-like grain structure (see optical micrograph in the insert to Fig. 1). The heat treatment which generated this grain structure (such that the grain size becomes equal to the wire diameter for better simulation of vacancy annihilation kinetics) and the quenching processes used are schematically depicted in Figure 1. Figure 1 also shows the solution treatment prior to quenching, conducted for 10 min at 570 °C. Quenching was performed either in water at room temperature (RT) (P1) or in a low-melting alloy at 160 °C (method described in Ref. [5]), with an isothermal hold of 120 s and subsequent cooling to RT in water (P2). For natural aging, samples were kept in a Peltier-cooled incubator at 25 °C.

Electrical resistivity measurements were carried out at −196 °C in liquid nitrogen using a custom-made four-point probe system and coiled wires (800 mm length) as samples. The current (NTN140–6.5ie high-precision source from FUG) and potential (2182A/E nanovoltmeter from Keithley) were measured separately at a constant applied current of 2 A. To minimize measurement errors, the change in specific resistivity resulting from natural aging $\Delta\rho = \rho_0 \times (R_a - R_0)/R_0$ was used [15,16], where ρ_0 is the as-quenched specific electrical resistivity, R_a is the resistance after natural aging and R_0 is the as-quenched electrical resistance of the coil.

Needle-shaped APT specimens were prepared via a standard two-step method [17]: after initial electropolishing of the wires with 10% perchloric acid and 90% methanol solution, 2% perchloric acid in butoxyethanol was used as the second electrolyte. APT was performed on a LEAP™ 4000X HR atom probe at a specimen temperature of 20 K and with a pulse fraction of 20%, a pulse rate of 200 kHz and a detection rate of 1% under ultrahigh-vacuum (<10^{-10} mbar) conditions. The software package IVAS 3.6.4™ from Cameca was deployed for the reconstruction procedure and analysis. Spatial distribution maps [18] were used to optimize the reconstruction parameters [19] in order to determine the correct crystallographic interplanar spacings (for Al) within the reconstructed data.

Simulation of excess-vacancy annihilation during the quenching processes P1 and P2 was performed using the FSAK model [13] as implemented in the thermokinetic simulation software package MatCalc [20,21]. With this model, the generation and annihilation of structural vacancies on ideal microstructural sources and sinks for vacancies, such as dislocation jogs, grain boundaries or Frank loops, can be predicted. Frank loops were not considered in the present work. Typical applications of this vacancy evolution model are reported, for example, in Refs. [22,23].

Figure 2 shows the change in specific electrical resistivity upon natural aging for the quenching procedures P1 and P2. For P1 an increase in electrical resistivity was observed. Such behavior is expected because solute clustering during natural aging is known to increase electrical resistivity [16]. The effect has been attributed to increased electron scattering at clusters [24] and depends in particular, but not solely, on the number density of solute clusters [25]. For Al–Mg–Si alloys, a significant increase in resistivity upon natural aging was reported [15,26], and the method has been utilized in recent studies [27,28] to monitor natural aging. The relatively small measured increase of 22 nΩ cm upon natural aging for 2.7×10^6 s after P1 quenching is associated with a low amount of Mg and Si in the current alloy and is comparable with the results for the lean Al–Mg–Si alloy studied in Ref. [4]. If quenching is, however, interrupted for 120 s according to P2, we find no increase in electrical resistivity in the timeframe of 2.7×10^6 s studied. Hence, we expect that P2 quenching suppresses the solute clustering during natural aging in the Al–Mg–Si alloy studied, at least for one month.

APT was used to investigate the distribution of solutes after natural aging for P1 and P2 quenching after

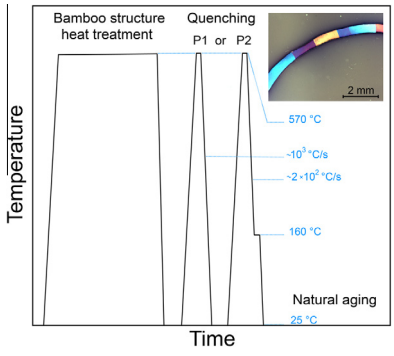

Figure 1. Schematic illustration of the heat treatment used to generate a controlled bamboo-like grain structure, and of the quenching processes P1 and P2. The resulting microstructure is shown in the insert.

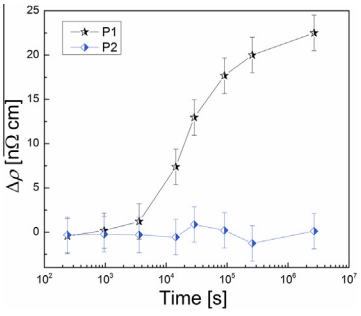

Figure 2. Evolution of the specific electrical resistivity change during natural aging at 25 °C after quenching processes P1 and P2.