Solute-vacancy clustering in Al-Mg-Si and Al-Si alloys

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Al-Mg-Si (6xxx series) aluminum alloys are in high demand as materials for vehicles because of their low weight, excellent formability and age hardenability. The usual process for heat treatment is solution heat treatment at approximately 820 K followed by quick quenching in water, resulting in a supersaturated solid solution (SSSS). After inevitable storage at room temperature (called natural aging, NA), the alloy is subjected to artificial aging (AA) at approximately 420 K, leading to a precipitation sequence: 11 SSSS \rightarrow Mg/Si/vacancy cluster \rightarrow Guinier Preston (GP) zone \rightarrow β " \rightarrow β " \rightarrow β (Mg₂Si).

It is well known that the early stage of solute clustering of Mg and Si proceeds quite quickly and is completed in less than an hour even at room temperature. A long-standing problem for the industry is that NA treatment often results in a negative effect on the mechanical strength in the subsequent AA treatment. From various studies on Al-Mg-Si alloys, vacancy behavior is considered to play an important role in the aging process, stimulating the diffusion of solute Mg and Si atoms and nucleation of clusters. Positron annihilation spectroscopy (PAS)^{3, 4)} and muon spin relaxation spectroscopy (μ SR)^{6, 7)} have been successfully used to investigate the vacancy and clustering behavior in Al-Mg-Si alloys.

New observations of time dependent muon spin relaxation and direct current (DC) magnetization of an Al-1.6%Mg₂Si alloy in the isothermal condition at 280, 290, or 300K are presented in this report. All the samples underwent heat treatment at 848 K for 1 h and subsequently quenching in ice water (STQ). Approximately 10 min after STQ, the sample was inserted into the ARGUS muon spectrometer, and then zero-field µSR measurement was started at a constant temperature. The observed spin relaxation spectra were fit using a Gaussian function with standard deviation 1/σ using the WIMDA program,⁸⁾ where σ is a measure of the depolarization rate of the muon spins as shown in Fig. 1(a). A similar sample treatment was performed in the magnetization measurements, which were carried out with a superconducting quantum interference (SQUID) magnetometer (Quantum Design, MPMS-XL7) (Fig. $1(b)^{9}$). The M_0 value is approximately $0.04 \text{ Am}^2/\text{kg}$. The time variations of the depolarization rates and the magnetizations at the corresponding measurement temperature appear to resemble each other. We attempted to estimate the migration energy of Mg/Si/vacancy in the non-equilibrium condition, assuming that the time variation of σ is brought about by the diffusion of these elements with the relations $\sigma = \sigma_0 \exp(-\tau t)$ and

 $\tau = \tau_0 \exp(-E/kT)$ (τ: reaction rate, and E: migration energy). From the recent studies of time dependent magnetizations⁹⁾ and positron annihilation spectroscopy³⁾ the Mg/Si/vacancy clustering (stage III) was found to proceed in a certain time window depending on the aging temperature. In the present σ vs. t curves we set the time windows as above 400, 70-500, and 30-150 min at 280, 290 and 300 K, respectively. The selected data points in Fig. 1(a) are marked with filled symbols. Fig. 2 shows an Arrhenius plot of $\ln(\tau)$ vs T which yields E = 0.18 eV. Further μSR experiments of Al-Mg-Si-Cu, Al-Si and Al-Mg samples have been conducted to investigate the details of solute elements and vacancy kinetics.

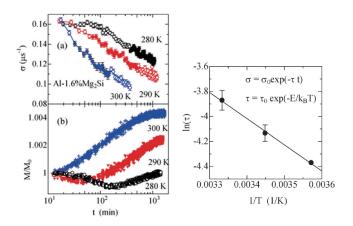


Fig. 1 (left). Aging time dependences of (a) zero-field spin relaxation rate and (b) normalized DC magnetization with an Al-1.6%Mg2Si sample at constant temperatures of 280, 290 and 300 K.

Fig. 2 (right). An Arrhenius plot for the reaction rate against temperature.

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