In situ three-dimensional investigation of hydrogen embrittlement in high strength aluminum alloys with modified precipitates

High-strength aluminum alloys are indispensable aerospace materials with superior strength-density ratios compared to other metal alloys. However, the development of ultrastrong commercial aluminum alloys has faced limitations in recent years owing to the strength-hydrogen embrittlement conflict. Hydrogen embrittlement, initially observed in iron and subsequently documented in various metallic materials, is particularly pronounced in high-strength aluminum alloys. Hydrogen-induced cracking is a direct result of the interactions of hydrogen atoms with diverse micro- and nanoscale structures. Although industrial over-aged aluminum alloys exhibit the ideal resistance to stress corrosion cracking, in which hydrogen embrittlement is the dominant mechanism, the precise role of precipitates in hydrogen trapping has only recently been discovered [1].

In this study, we demonstrate the strategic transformation of nanosized age-hardening precipitates, which are commonly used as core strengthening elements in high-strength aluminum alloys, into potent hydrogen trap sites. In contrast to the problematic η precipitates, collectively termed η-$MgZn_2$, which pose risks of interfacial debonding [2], we highlight the highly effective hydrogen trapping capability of T precipitates ($Al_2Mg_3Zn_3$). Our investigation assessed the effectiveness of these precipitates in mitigating hydrogen embrittlement and the related mechanisms, using a typical Al-Mg-Zn-Cu aluminum alloy as a model material.

Quaternary 7XXX aluminum alloys with a chemical composition of Al-5.6Zn-2.5Mg-1.6Cu (mass%) were prepared to induce a partial switch from the η phase to the T phase through adjustments to the

aging parameters. Elevating the aging temperature from low temperature (LT) to high temperature (HT) facilitated a transformation from η to T, validated by scanning transmission electron microscopy (STEM) and diffraction patterns. The microstructure observed in Fig. 1(a) confirms the significant presence of the T phase in the HT material. Fig. 1(b) illustrates the results from the first-principles simulations, showing the exceptional hydrogen trapping capacity within the interior of the T precipitates with a maximum binding energy of 0.6 eV.

In situ tensile tests under synchrotron X-ray tomography were performed at the undulator beamline SPring-8 **BL20XU** with a beam energy of 20 keV. Significantly improved ductility owing to the change in the nanoprecipitates is shown in Fig. 2. The four-dimensional observations demonstrated a reduced growth rate of the main crack in the presence of the T phase. In contrast to the hydrogen-induced grain boundary (GB) separation in the LT material, the main crack in the T-phase-rich material remained almost stagnant with increasing applied strain until the final fracture.

According to the decohesion mechanism [3], hydrogen reduces the energy required to separate various interfaces, and hydrogen coverage at the grain boundaries controls the growth behavior of intergranular cracks. The intense stress field in the vicinity of the GB ahead of the crack tip attracted more hydrogen atoms toward it, and the crack growth speed was significantly affected by the initial hydrogen occupancy at the GBs. Moreover, other mechanisms, particularly hydrogen-enhanced local plasticity [4], can also facilitate intergranular fracture owing to the
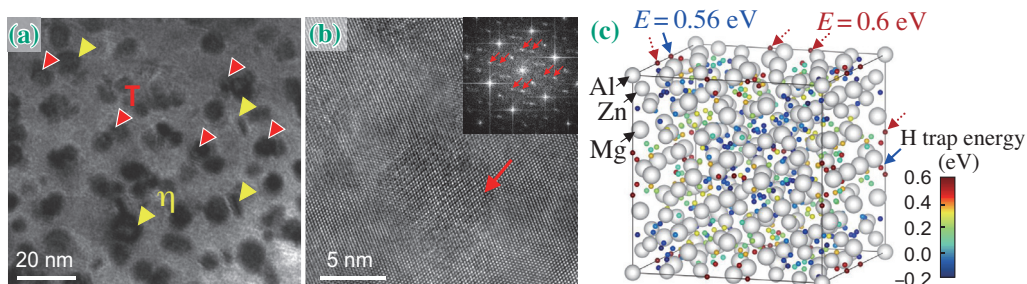

Fig. 1. (a) and (b) TEM images and fast Fourier transform analysis of T phase precipitates in HT materials. (c) Distribution of hydrogen trapping energies in the interior of the T phase with a maximum energy of 0.6 eV predicted by first-principles calculations.

dislocation interaction with GBs, which alters the local stress and strain states, GB structure, and hydrogen distribution through dislocation accommodation, pile-up, and penetration through GBs.

The presence of numerous nanosized T precipitates is expected to result in diminished hydrogen coverage at the dislocations and GBs near the crack tip. This reduction is expected to weaken the hydrogen-enhanced local plasticity, and consequently mitigate its contribution to hydrogen-induced debonding at the GBs. Therefore, the cohesive energy of the GB can be maintained at a sufficiently high level, surpassing the critical value required for separation.

Quantitative assessments of the hydrogen concentration and occupancy at the dislocations, GBs, vacancies, precipitates, particles, and microvoids within the material were conducted through local partitioning calculations. In the absence of neighboring voids, the hydrogen trapping effect in the T phase is estimated

to result in a substantial reduction of 2−3 orders of magnitude in the hydrogen concentrations at dislocations, GBs, and vacancies. This reduction is indicative of the robust hydrogen trapping effect of the T phase [5].

A competition effect arises between two nanoprecipitate types: hydrogen trapping at coherent η/Al interfaces induces interfacial debonding, while hydrogen trapping within the T phase effectively suppresses it. The role of the T phase in hydrogen embrittlement suppression is attributed to its robust hydrogen-trapping capacity and stress localization mitigation. Our novel strategy modifies nanoscopic precipitates, transforming them into potent hydrogen traps. This strategy is expected to be universally effective for various high-strength aluminum alloys because of the widespread availability of the T phase, which may inspire the development of hydrogen-resistant alloys with similar switchable nanostructures.
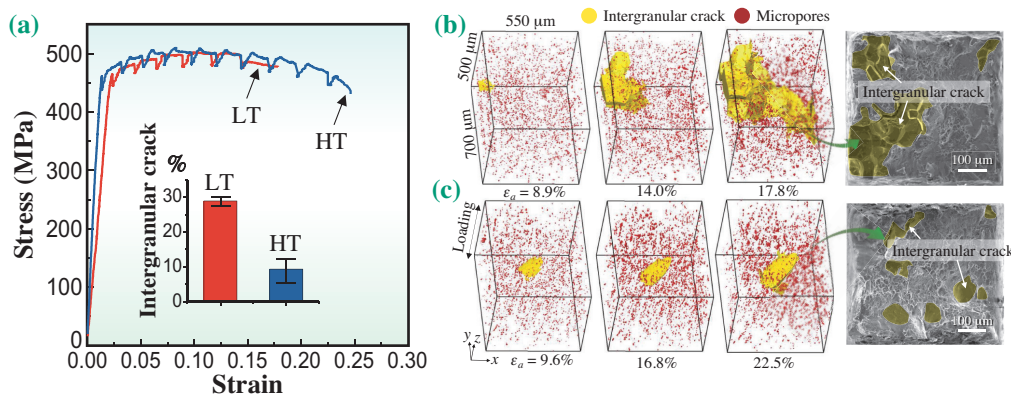

Fig. 2. (a) Stress–strain curves for LT and HT materials, with the inset figure showing the average areal fractions of intergranular cracks (IGCs) on the fracture surfaces. The 3D renderings of the IGCs and corresponding fracture morphologies in LT and HT materials are shown in (b) and (c), respectively.

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