Scripta Materialia 135 (2017) 19–23

Contents lists available at ScienceDirect

Scripta Materialia

journal homepage: www.elsevier.com/locate/scriptamat

Regular Article

Influence of intermetallic particles on the initiation and growth behavior of hydrogen micropores during high-temperature exposure in Al-Zn-Mg-Cu aluminum alloys

Hang Su [a,*], Shahnewaz Bhuiyan [a], Hiroyuki Toda [a], Kentaro Uesugi [b], Akihisa Takeuchi [b], Yoshio Watanabe [c]

[a] Department of Mechanical Engineering, Kyushu University, Japan
[b] Japan Synchrotron Radiation Institute (JASRI), Japan
[c] UACJ Corporation, Japan

ARTICLE INFO

Article history:
Received 12 January 2017
Received in revised form 13 March 2017
Accepted 13 March 2017

Keywords:
X-ray microtomography
Synchrotron radiation
Hydrogen micropores
Al-Zn-Mg-Cu aluminum alloy

ABSTRACT

X-ray tomography is employed to observe the effects of intermetallic compound particles on the nucleation and growth of hydrogen micropores at high temperatures in Al-Zn-Mg-Cu aluminum alloys. Hydrogen micropores are heterogeneously nucleated on particles during exposure at 748 K. Growth and coalescence of the hydrogen micropores are observed with increasing exposure time. Interactions between hydrogen micropores and particles have a significant influence on the growth and coalescence of hydrogen micro pores. The growth speed of hydrogen micropores, nucleated on spherical, small particles is faster than those on other nucleation sites.

Recently, a series of research studies have been performed to improve the strength and fracture toughness of Al-Zn-Mg-Cu aluminum alloys [1–3]. However, increasing strength leads to an increase in hydrogen embrittlement sensitivity of the precipitated strengthened aluminum alloys [4]. Young, et al. have revealed that the growth rate of hydrogen-induced cracks under the peak-aged condition is around twenty times higher than that of under-aged conditions in 7050 aluminum alloys [5]. Bhuiyan, et al. have compared the hydrogen-induced quasi-cleavage fracture behavior of Al-Zn-Mg-Cu aluminum alloys with different Zn contents, revealing that the hydrogen susceptibility is increased with the increase in Zn content [6].

A great number of hydrogen atoms are absorbed into aluminum alloys due to the destruction of the oxide film during high-temperature heat treatment, and the content of hydrogen is thereby much higher compared to the hydrogen solubility at room temperature [7,8]. With the help of thermal desorption spectroscopy, it has been clarified that supersaturated hydrogen atoms in aluminum alloys are partitioned to trap sites such as interstitial lattices, vacancies, dislocations, solute atoms, precipitates, intermetallic particles and high-angle grain boundaries [9–12]. Hydrogen micropores are characterized as one of the hydrogen trap sites in aluminum alloys [9,13,14]. Toda, et al. have revealed that over 53% of hydrogen atoms are sometimes trapped in micropores in Al—Mg aluminum alloys [13]. The initiation and growth of hydrogen micropores at high temperatures have been studied by several researchers in recent years. By applying first-principles calculations, Liu, et al. have revealed that hydrogen micropores are formed from hydrogen-vacancy clusters [15]. On the other hand, Toda, et al. have revealed with the help of X-ray tomography that hydrogen micropores are heterogeneously initiated on the intermetallic particles in Al—Mg aluminum alloys. The growth of hydrogen micropores nucleated on particles is attributed to the high internal hydrogen pressure-induced creep deformation of the surrounding aluminum alloys [13]. In addition, hydrogen micropores nucleated on particles show premature growth under load, and approximately 7–28% of the dimples on the fracture surface originate from hydrogen micropores [16].

Intermetallic compound particles, such as Al_7Cu_2Fe, Mg_2Si and Al_2CuMg, are observed in Al-Zn-Mg-Cu aluminum alloys [17–20]. According to previous research, all of these particles can be characterized as heterogeneous nucleation sites of hydrogen micropores during high-temperature exposure [9,13,21]. Recently, Bhuiyan, et al. have revealed that approximately 51% of hydrogen is trapped into hydrogen micropores nucleated on particles in Al-Zn-Mg-Cu aluminum alloys, indicating their importance on hydrogen partitioning behavior [9]. On the other hand, hydrogen micropores initiated on the particles might influence the damage evolution at the particles during loading. Su, et al. have revealed that hydrogen micropores initiated on Al_7Cu_2Fe particles accelerate the formation of micro cracks under load through linkage with

* Corresponding author.
E-mail address: suzo1221@gmail.com (H. Su).

http://dx.doi.org/10.1016/j.scriptamat.2017.03.020
1359-6462/

newly formed voids due to particle fracture [21]. In the present research, the aim is to reveal the formation behaviors of hydrogen micropores in a practical Al-Zn-Mg-Cu aluminum alloy containing various intermetallic particles during high-temperature exposure. High-resolution X-ray tomography, which enables the visualization of the initiation, growth and annihilation of each micropore, is applied to clarify the above issue.

An as-cast Al-Zn-Mg-Cu aluminum alloy was used in the present research; its chemical composition was 10.00 Zn, 2.40 Mg, 1.50 Cu, 0.30 Fe, 0.30 Si, 0.15 Zr, 0.04 Ti and balance Al in mass %. To reveal the influence of intermetallic particles on the initiation and growth of hydrogen micropores during a homogenization treatment, ex-situ heating experiments were performed on the same specimen at 748 K for exposure times between 0 and 145.2 ks. A specimen 10 mm in length and 0.6×0.6 mm in cross-section was used for the X-ray tomography observation. A region 300 μm in height and 300×300 μm in cross section was extracted to investigate the nucleation sites of the hydrogen micropores.

The X-ray tomography experiment was performed at the BL20XU beamline in SPring-8. A monochromatic X-ray beam with a photon energy of 20 KeV, generated by a liquid nitrogen-cooled Si (111) double crystal monochromator, was applied for the 3D observations. An image detector was located 20 mm behind a specimen. The image detector consisted of a 4000 (H) $\times$ 2624 (V) element CMOS camera, a single-crystal scintillator (Lu_2SiO_5: Ce) and a lens (20$\times$). Eighteen hundred radiographs with scanning of 180°, were captured in 0.1° increments. The entire cross-section and an approximately 1024 μm region of the specimen were captured on the CMOS camera. Image slices were reconstructed by the conventional filtered back-projection algorithm. The linear absorption coefficient (LAC) of $-30-40$ cm^{-1} fell within an 8-bit grayscale from 0 to 255 for a conversion process to 8-bit. The isotropic voxels in the reconstructed images were $(0.5\ \mu m)^3$ in size.

Fig. 1 shows the 3D reconstructed images of the same specimen, including hydrogen micropores and intermetallic particles that are homogenized for different exposure times in Al-Zn-Mg-Cu aluminum alloys. The majority of the Al_2CuMg, Al_7Cu_2Fe and Mg_2Si particles are located along the grain boundaries in the as-cast Al-Zn-Mg-Cu aluminum alloys. The LAC values of the Mg_2Si, Al_2CuMg and Al_7Cu_2Fe particles range from -15 to 2, 30–40 and 27–35 cm^{-1}, respectively. The Cu-bearing particles are hereinafter expressed as Al_2CuMg/Al_7Cu_2Fe due to the overlap of the LAC values. With increasing exposure time, Al_2CuMg/Al_7Cu_2Fe particles are gradually dissolved into the matrix while Mg_2Si particles remain in the materials. Deng, et al. have studied the transformation of intermetallic particles during the homogenization process in Al-Zn-Mg-Cu aluminum alloys. After being homogenized at 743 K for more than 24 h, Al_2CuMg particles dissolve into the matrix, and only irregular Al_7Cu_2Fe particles remain along the grain boundaries [18]. Although it was impossible to separate Al_2CuMg and Al_7Cu_2Fe particles in the present work, it can still be inferred that only Al_7Cu_2Fe particles remain in the matrix after long exposure.

The initiation, growth and annihilation of hydrogen micropores during high-temperature exposure can also be seen in Fig. 1. In the as-cast condition, shrinkage cavities with complex shapes are seen to be located at the interface between the intermetallic particles and the matrix. During the homogenization treatment at 748 K for 0.8 ks and 1.9 ks, the shrinkage cavities gradually become spherical, as shown in pore A in Fig. 1a–c. Instead, small hydrogen micropores are heterogeneously nucleated at high density on the intermetallic particles, as shown in Fig. 1b–c, making the spatial distribution of hydrogen micropores relatively homogeneous. With the increase in exposure time, the hydrogen micropores continue to grow, as shown in Fig. 1d–f. During the homogenization treatment at 748 K, pore A gradually grows and then coalesces with neighboring hydrogen micropores after being exposed for 4.5 ks. Pore B shows an independent growth without coalescence up to 145.2 ks. Pore C is initiated on Al_2CuMg/Al_7Cu_2Fe particles at 4.5 ks and annihilates after being exposed for 145.2 ks. According to Toda, et al., Ostwald ripening is the growth mechanism of hydrogen micropores, and small hydrogen micropores gradually annihilate with the increase in exposure time [13]. It is worth noting that hydrogen micropores exhibit octahedral shapes consisting of eight {111} planes that are energetically stable in aluminum alloys [13] after being exposed to 748 K for 145.2 ks, as shown in Fig. 1f.

Fig. 2 shows cross-sectional images, showing intermetallic particles and hydrogen micropores that are observed after being exposed at 748 K for 10.8 ks, 25.8 ks, 61.2 ks and 145.2 ks. For the irregular, large Al_2CuMg/Al_7Cu_2Fe particles, hydrogen micropores tend to be initiated on the tips of these particles, as shown as particle A in Fig. 2d. With the increase in exposure time, particles gradually dissolve into the matrix, and hydrogen micropores continue to grow. The coalescence of hydrogen micropores is observed after being exposed at 748 K from 25.8 ks to 145.2 ks, as shown as pores A, B and C in Fig. 2a. Closely spaced

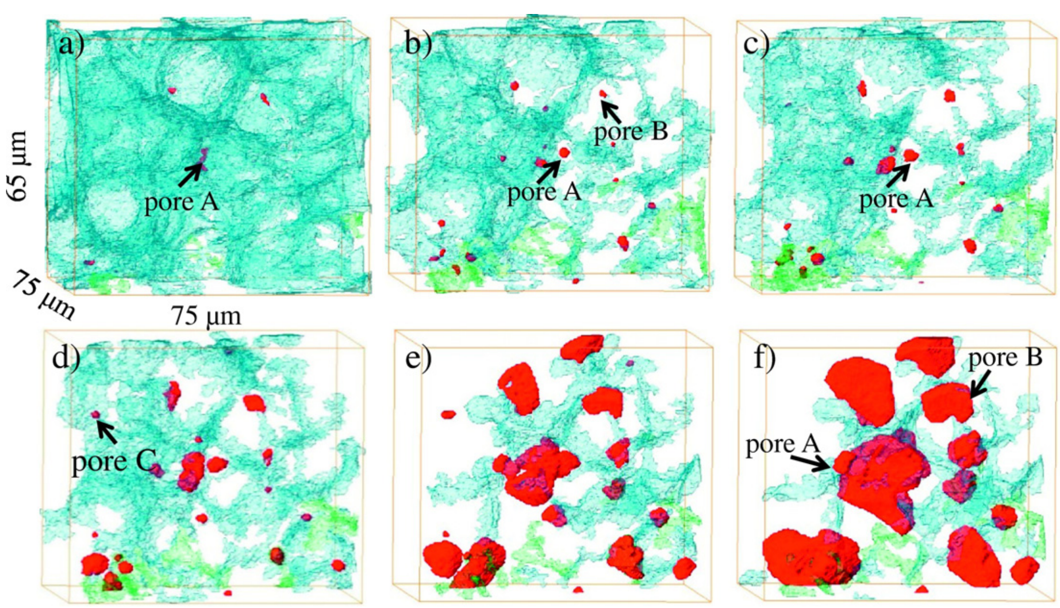

Fig. 1. 3D rendered images of an identical region in the material exposed at 748 K for: a) 0 ks, b) 0.8 ks, c) 1.9 ks, d) 4.5 ks, e) 25.8 ks f) 145.2 ks. Hydrogen micropores are shown in red, Al2CuMg/Al7Cu2Fe particles are shown in blue and Mg2Si particles are shown in green.