Brittle-to-ductile transition in polycrystalline aluminum containing gallium in the grain boundaries

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Brittle-to-ductile transition in polycrystalline aluminum containing gallium in the grain boundaries

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Abstract. It is well known that aluminum/gallium couple causes liquid metal embrittlement. Gallium atoms penetrate the grain boundaries of polycrystalline aluminum and degrade it. Polycrystalline aluminum specimens were contacted with a small droplet of gallium for 24 h. After gallium was removed from the surface of the specimens, tensile tests were performed between 77 K and 313 K. The specimens are ductile below 230 K and brittle above 303 K, the melting temperature of gallium. Between 280 K and 300 K, the maximum stress is larger in the specimens heated from 77 K than in those cooled from 313 K. This thermal history dependence of the maximum stress is considered to be attributed to the solidification of supercooled gallium in the grain boundaries.

1. Introduction

Polycrystalline aluminum becomes brittle if it is stressed in liquid gallium. This is a typical example of liquid metal embrittlement, which refers to the loss of ductility of normally ductile metals when stressed whilst in contact with a liquid metal. In the case of aluminum and gallium, it is also known that fracture stress and fracture strain are reduced above the melting point of gallium (29.8°C), even if it is stressed after gallium is removed from the surface of aluminum specimens. The fracture stress reduces with an increase in the contact time. This phenomenon is attributed to the penetration of gallium into the polycrystalline aluminum, which reduces the strength of grain boundaries [1]. The rapid penetration of gallium atoms has been confirmed by radiotracer [2], transmission electron microscope [3–5], and X ray techniques [6, 7].

Polycrystalline aluminum containing gallium in the grain boundaries shows a large ultrasonic attenuation above the melting temperature of gallium, while the attenuation is nearly the same as in polycrystalline aluminum without gallium at low temperatures [8]. A change in ultrasonic attenuation occurs over a small temperature range, and the temperature depends on the thermal history. The change in the ultrasonic attenuation may be accompanied by changes in some mechanical properties of the same specimen. In this paper, we describe the fracture properties of polycrystalline aluminum containing gallium between 77 K and 313 K.

2. Experimental

Specimens were spark cut from a 3 mm thick polycrystalline aluminum plate to the dimensions shown in figure 1. The purity of the specimens was 99.999% and the average grain size was 0.1 mm. After the specimens were chemically polished and annealed at 700 K, a small drop of liquid gallium saturated with aluminum was placed on the surface of the specimens at 313 K. To ensure a contact between aluminum and liquid gallium, an aluminum surface covered with the liquid gallium was scratched with a needle. After keeping the specimens at 308 K for 24 h, they were immersed in liquid nitrogen and then gallium was removed from the surface with emery paper. Since the penetration velocity of gallium atoms into the grain boundaries of polycrysltalline aluminum is several micrometers per second [1, 2, 4], 24 h is long enough for the grain boundaries to be invaded by gallium over the entire thickness of the specimen.

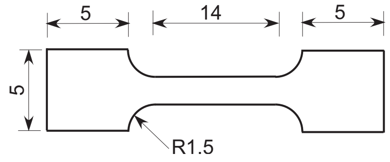

Figure 1. Tensile specimen with thickness of 3 mm.

Tensile tests were performed at a strain rate $\dot{\epsilon}$ of 1.2×10^{-2}/s or 1.2×10^{-3}/s. Two types of thermal histories were examined. Some of the specimens were immersed in ethanol at 313 K and then cooled to the test temperature, and the others were immersed in liquid nitrogen and then heated to the test temperature. The fracture surface was examined using a scanning electron microscope.

3. Results

The typical stress strain curves obtained at $\dot{\epsilon} = 1.2\times10^{-2}$/s are shown in figure 2. The nominal stress and nominal strain are shown in this and the following figures. At 313 K, both the maximum stress $\sigma_{\max}$ and the fracture strain ϵ_f are considerably reduced by gallium, while at 77 K, $\sigma_{\max}$ is not affected, but ϵ_f is somewhat reduced. At 281 K, $\sigma_{\max}$ is larger in the specimen heated from 77 K than that cooled from 313 K. At this temperature, both $\sigma_{\max}$ and ϵ_f depend on the thermal history. At 242 K, $\sigma_{\max}$ is almost the same for the heated and cooled specimens; however, ϵ_f seems to depend on the thermal history.

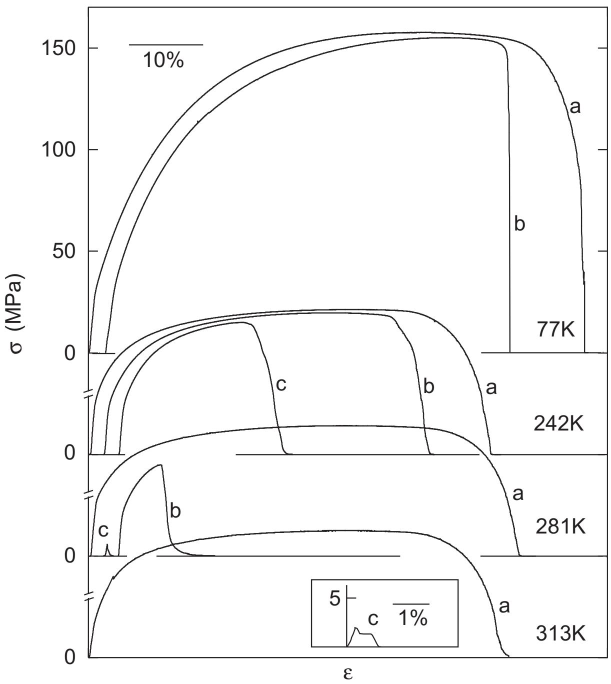

Figure 2. Stress strain curves of polycrystalline aluminum without gallium (a) and with gallium in the grain boundaries stressed after heating from 77 K (b) and cooling from 313 K (c). Temperatures indicated in the figure are the test temperatures.

The temperature dependence of $\sigma_{\max}$ and ϵ_f measured at $\dot{\epsilon} = 1.2 \times 10^{-2}$/s are shown in figure 3. The broken curves represent $\sigma_{\max}$ and ϵ_f of polycrystalline aluminum without gallium. Both $\sigma_{\max}$ and ϵ_f decrease with an increase in temperature. The effect of gallium is noticeable in $\sigma_{\max}$ above 230 K. The maximum stress $\sigma_{\max}$ of the specimens heated from 77 K decreases gradually, and above 300 K, it is about the same as that measured in a polycrystalline aluminum specimen stressed in liquid gallium. On the other hand, $\sigma_{\max}$ of specimens cooled from 313 K shows an abrupt increase at 280 K. Between 300 K and 280 K, $\sigma_{\max}$ depends on the thermal history. The fracture strain ϵ_f also decreases with temperature; however, the hysteresis is less clear because of the scattering of data.

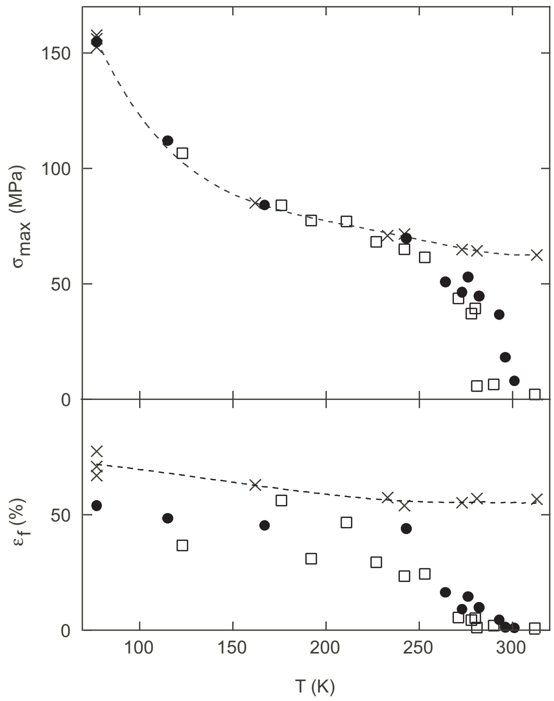

Figure 3. Maximum stress $\sigma_{\rm max}$ (upper) and fracture strain $\epsilon_{\rm f}$ (lower) as functions of measuring temperature. $\dot{\epsilon} = 1.2 \times 10^{-2}$/s.
$\times$ polycrystalline aluminum without gallium.
● polycrystalline aluminum with gallium in the grain boundaries heated from 77 K.
□ polycrystalline aluminum with gallium in the grain boundaries cooled from 313 K.

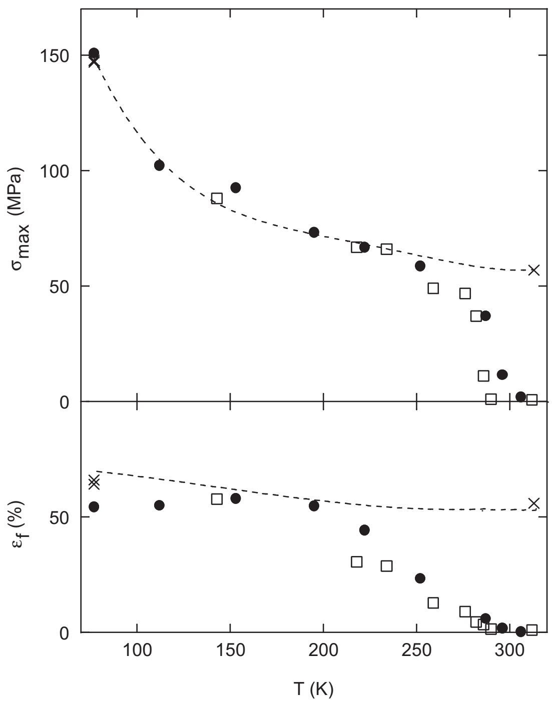

Figure 4. Maximum stress $\sigma_{\rm max}$ (upper) and fracture strain $\epsilon_{\rm f}$ (lower) as functions of measuring temperature. $\dot{\epsilon} = 1.2 \times 10^{-3}$/s. Symbols are the same as those shown in figure 3.

Figure 4 shows $\sigma_{\rm max}$ and $\epsilon_{\rm f}$ obtained at $\dot{\epsilon} = 1.2 \times 10^{-3}$/s. Similar tendencies as shown in figure 3 are observed for both $\sigma_{\rm max}$ and $\epsilon_{\rm f}$. The maximum stress $\sigma_{\rm max}$ of the specimens cooled from 313 K changes abruptly around 285 K, which is higher by 5° than that shown in figure 3.

Figure 5 shows the SEM of fracture surfaces. When the maximum stress is low, most of the fracture surface consists of smooth areas, suggesting intergranular fracture. When the maximum stress is large, the area of transgranular fracture increases. Secondary cracks are often observed perpendicular to the fracture surface.

4. Discussion

Since aluminum specimens without gallium are ductile in all the temperature range tested, transition from ductile fracture at low temperatures to brittle fracture at high temperatures observed in the specimens with gallium is attributed to gallium in the grain boundaries of polycrystalline aluminum.

The degradation consists of two components: a gradual decrease in $\sigma_{\rm max}$ above 230 K, and very small $\sigma_{\rm max}$ above 280 K or 285 K, which is only observed in the specimens cooled from above the melting temperature of gallium. The second component is, therefore, considered to be attributed to supercooled Ga in the grain boundaries, and an abrupt change in $\sigma_{\rm max}$ is caused by the solidification of supercooled gallium. Although at atmospheric pressure, stable α-gallium melts at 303 K, gallium can be easily supercooled, especially in small particles [9].

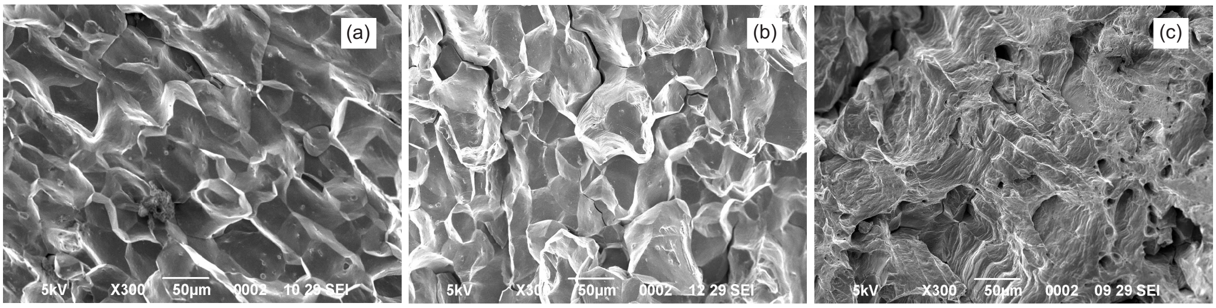

Figure 5. SEM of fracture surfaces of (a) a polycrystalline aluminum specimen cooled from 313 K and tensile tested at 281 K, (b) a specimen heated from 77 K and tested at 281 K, (c) a specimen heated from 77 K and tested at 242 K. The strain rate was 1.2×10^{-2}/s.

The thickness of gallium layers in some grain boundaries is measured to be of the order of 1 μm [6], and gallium in these grain boundaries may have similar properties as small particles have. Ultrasonic attenuation measured in polycrystalline aluminum containing gallium shows an abrupt decrease in attenuation around this temperature when the specimen is cooled from 308 K, while the attenuation is small up to 300 K when the specimen is heated from 200 K [8]. This temperature dependence of the ultrasonic attenuation also supports the solidification of supercooled gallium. The transition temperature of the ultrasonic attenuation and the abrupt change in $\sigma_{\max}$ cannot be compared directly, because both temperatures depend on various factors such as content of gallium, stress and changing rate of temperature.

Between 230 K and 280 K, a gradually decreasing component of $\sigma_{\max}$ is independent of the thermal history. SEM micrographs show that most of the fracture surface consists of smooth grain surfaces when the maximum stress is small, while the fracture surface is mixture of intergranular and transgranular fracture surfaces when the maximum stress in large. This suggests that in this temperature range the number of weakened grain boundaries increases with temperature, even when gallium in the grain boundaries may not be in the liquid state. The penetration of gallium depends on the nature of grain boundaries of aluminum [5]; therefore, it can be expected that each grain boundary becomes weak at a different temperature.

Penetration of liquid metal atoms into the grain boundaries of a solid metal has been confirmed in several solid metal and liquid metal systems which cause liquid metal embrittlement, such as copper/bismuth [10] and silver/mercury [1] systems. In these systems the similar brittle-to-ductile transition may be observed near the melting point of the liquid metal.

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