Recovery of ductility observed in liquid gallium induced embrittlement of polycrystalline silver

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Abstract Polycrystalline silver is embrittled by liquid gallium immediately after intimate contact between the solid and the liquid metals is made. When the tensile test is performed at room temperature, recovery of ductility has been observed if the contact time is long enough. This recovery is induced by formation of an intermetallic compound between silver and gallium. When the specimen is stressed above 420 K, embrittlement is not observed even if the contact time between the solid and liquid metals is short. This is the ductility trough reported in various solid and liquid metal couples that cause the liquid metal embrittlement. Although loss of embrittlement occurs as a function of contact time and as a function of test temperature, it may be difficult to connect the ductility trough with contact time dependent recovery from embrittlement, and there seems two independent recovery processes.

Keywords liquid metal embrittlement, ductility trough, sliver, gallium

1. Introduction

Liquid metal embrittlement (LME) is a phenomenon that normally ductile metals become brittle when they are stressed in close contact with certain kinds of liquid metals. Embrittlement appears as a decrease in maximum stress and fracture strain in a tensile test. Characteristic features of LME are [1]:
(1) LME occurs only in some solid and liquid metal couples. This is called “selectivity” and solid and liquid metal couples that cause LME are listed in ref. [2]. Solid and liquid metal couples such as polycrystalline aluminum and liquid gallium show a significant reduction of the maximum stress and fracture strain, while in case of a steel and liquid lead the maximum stress decreases to only a half of the original maximum stress [3]. The factors that determine the selectivity are not known.
(2) The embrittlement takes place immediately after intimate contact between solid and liquid metal is made. For polycrystalline aluminum-liquid gallium and polycrystalline silver-liquid mercury couples, embrittlement occurs even after the surface liquid metal is removed before stressing, if the contact time of the solid and liquid metals is long enough. In these cases, the degree of embrittlement increases with the contact time after a certain incubation time [4].
(3) The degree of embrittlement is severe near the melting temperature of the liquid metal, and the embrittlement disappears at a higher temperature. This temperature range is called “ductility trough”, and is often explained as a result of temperature dependence of the yield stress [3] or stress relaxation at potential crack initiation site [5].

We have reported that liquid gallium induced embrittlement of polycrystalline silver is transitory [6, 7]. The silver tensile specimens with a small amount of gallium on the surface were kept in a furnace at the temperature T_h for a period of time t_h. The tensile test was performed to the specimens at 308 K. When t_h is small, the specimens undergo severe reduction of maximum stress and fracture strain, while for large t_h, the specimens show ductile behavior. The duration of brittleness is short when the quantity of gallium is small and the temperature of the furnace is high. For silver-gallium couple ductility trough exists as a function of the contact time.

The objective of the present investigation is to show that the ductility trough exits as a function of temperature as many other embrittlement couples, and to discuss differences between the ductility

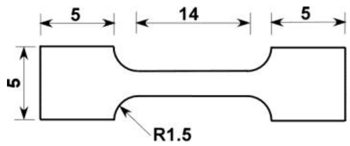

Fig. 1 Tensile specimen. The thickness is 2 mm.

trough as a function of temperature and that as a function of time.

2. Experimental

The tensile specimens used in this investigation is shown in Fig. 1. They were cut out from a silver plate of 99.98 % purity by electrodischarge machining. After mechanical polishing, the specimens were annealed at 1073 K for 2 hours. The average grain size of the specimens was 0.3 mm. A small amount of liquid gallium, 10 mg in weight and saturated with silver at 308 K, was deposited on the specimen, so that one of the four faces of the gauge section was completely covered with the gallium. To ensure the contact between the solid specimen and the liquid, the surface covered with liquid gallium was scratched with a needle. After immersing the specimen in liquid nitrogen to freeze the gallium, the specimen was placed in the tensile machine. The tensile test was performed on the specimen in a hot silicone oil bath in the temperature between 300 K and 465 K and in water between 274 K and 300 K. The strain rate was 1.2×10^{-2} s^{-1}.

The fracture surface was observed by the scanning electron micrograph (SEM).

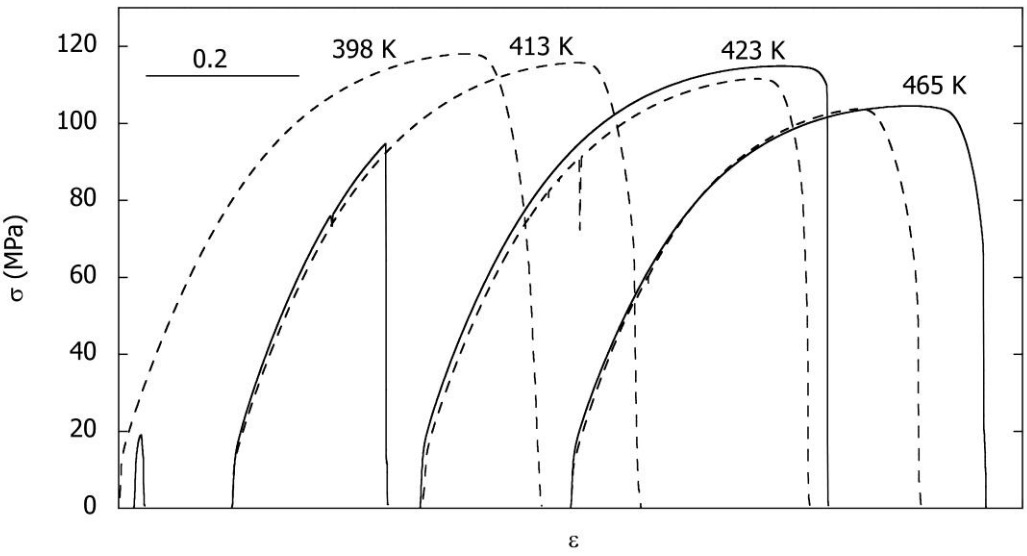

Figure 2. Stress-strain curves of silver specimen with gallium (solid lines) and without gallium (dashed lines). The test temperature is shown in the figure.

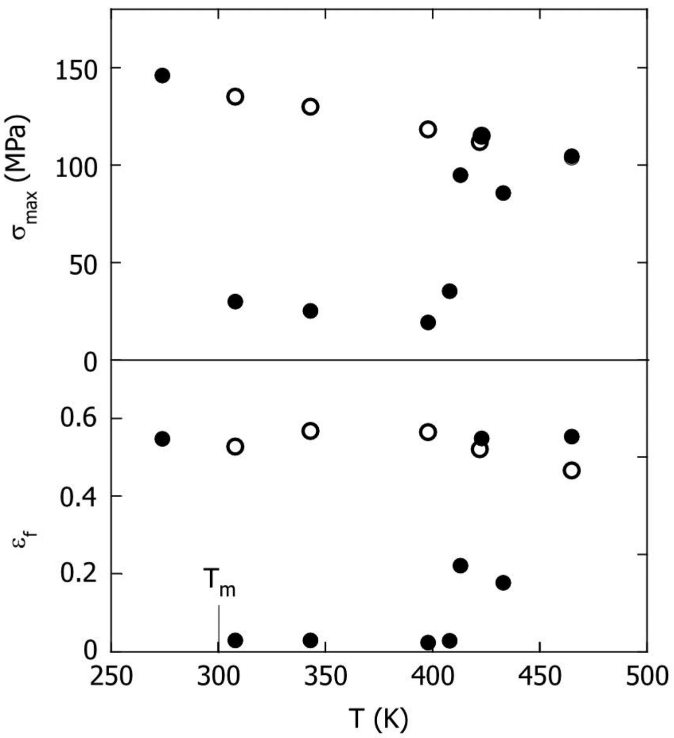

Figure 3. Temperature dependence of maximum stress σ_{max} and frcture strain ε_f of silver specimens tested with liquid gallium (solid circles) and without gallium (open circles). T_m indicates the melting point of gallium.

3. Results

Figure 2 shows some typical stress-strain curves of the specimens without and with liquid gallium on the surface. The test temperature is shown in the figure. Below 400 K, liquid gallium induces a significant reduction of the maximum stress σ_{max} and the fracture strain ε_f, while at 465 K liquid gallium has no effect on σ_{max} and no reduction of ε_f is observed. Around the transition temperature (413 K and 423 K in the figure), in the specimens with gallium the stress drops abruptly after the stress-strain curve reaches the maximum, while the decrease of the stress is gradual in the specimens without gallium at all temperatures measured and in the specimen with gallium at 465 K.

In Fig. 3 the maximum stress σ_{max} and the fracture strain ε_f are shown as a function of test temperature *T*. When the specimen is pulled without gallium, the maximum stress decreases with temperature, while the fracture strain is almost constant. Since the specimen was immersed in liquid nitrogen before the test to prevent supercooling of gallium, embrittlement is not observed below the melting temperature T_m of gallium (303 K). Above T_m the maximum stress and the fracture strain are reduced significantly by liquid gallium. The brittleness disappears above 420 K. This means that ductility trough starts at the melting temperature of gallium and ends around 420 K.

Figure 4 shows the SEM images of the fracture surfaces and the optical micrographs of the tensile specimens. When the test temperature is below 413 K, the fracture surface is apparently intergranular, although the surface is partially covered with liquid gallium, and the optical micrograph shows that the fracture surface is nearly perpendicular to the tensile axis. At 465 K, necking is observed in the specimen. The fracture surface is covered with liquid gallium. This

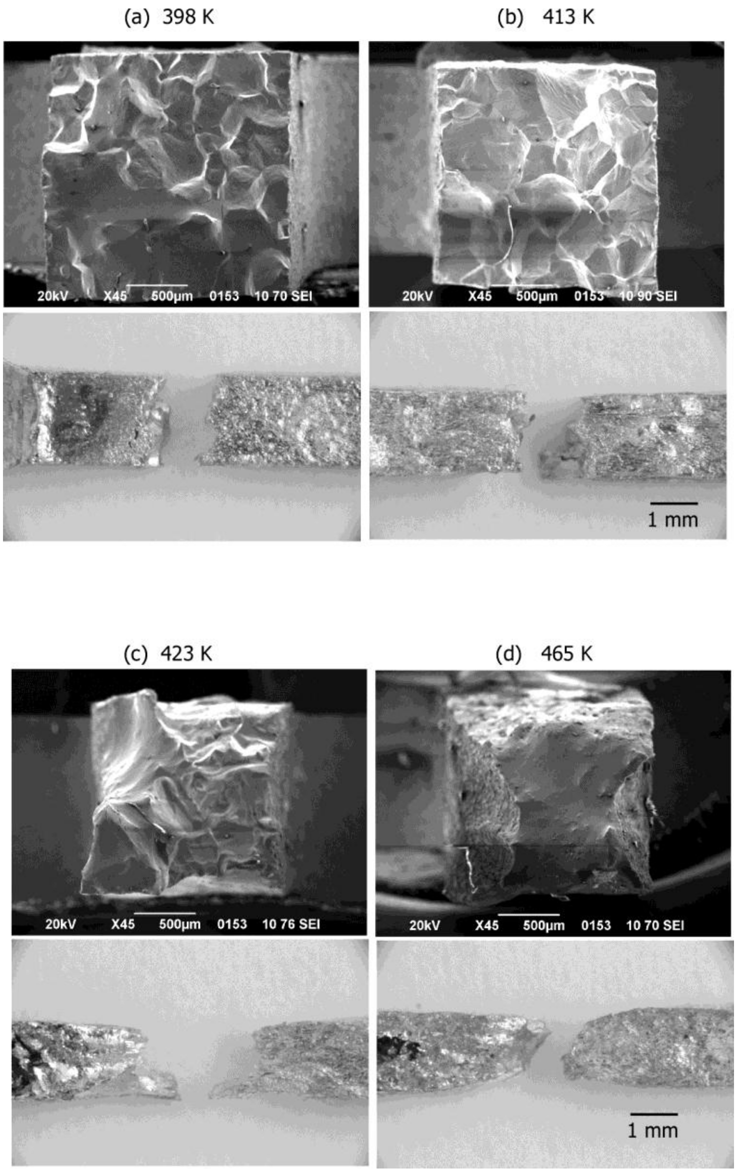

Figure 4. Scanning micrograph of the fracture surface (upper) and optical micrograph of the specimen (lower) tested at (a) 398 K, (b) 413 K, (c) 423K and (d) 465 K.

indicates that gallium is in liquid phase when the specimen fails. At 423 K, in spite of large maximum stress and fracture strain, the optical micrograph shows that reduction of cross-sectional area does not localize and the fracture surface is rough, which is different from the typical ductile fracture surface.

4. Discussion

In Fig. 3 we showed that the ductility trough exists as a function of temperature in polycrystalline silver-liquid gallium couple.

We have reported in [7] that for this couple the recovery from LME is observed if the contact time between the solid and liquid metals is long. In [7] the tensile test was performed at 308 K on the specimens with 10 mg of gallium after they were kept in a furnace at the temperature T_h for certain holding times t_h. Stress-strain curves are shown in Fig. 5(a) for the specimens of T_h =373 K. In Fig.

5(b) the maximum stress is shown as a function of the holding time t_h. Ductility recovers around 80 ks. Figure 5(c) shows the recovery time t_R as a function of the furnace temperature T_h. The recovery observed in the time dependence is considered due to formation of ζ'-phase or Ag_7Ga_3 by the X-ray diffraction analysis.

From Fig. 5(c), the recovery time of the specimen held at 465 K is estimated more than 10 ks, when it is stressed at 308 K. In the present experiment, however, the specimens stressed at 465 K is not embrittled, although the test was performed immediately after the specimen temperature became the test temperature, and the test ended in less than a few minutes. The specimen that is ductile at 465 K becomes brittle at 308 K. As shown in Fig. 4(d), the fracture surface of the specimen pulled at 465 K is covered with gallium flowing from the specimen surface. At this temperature the specimen

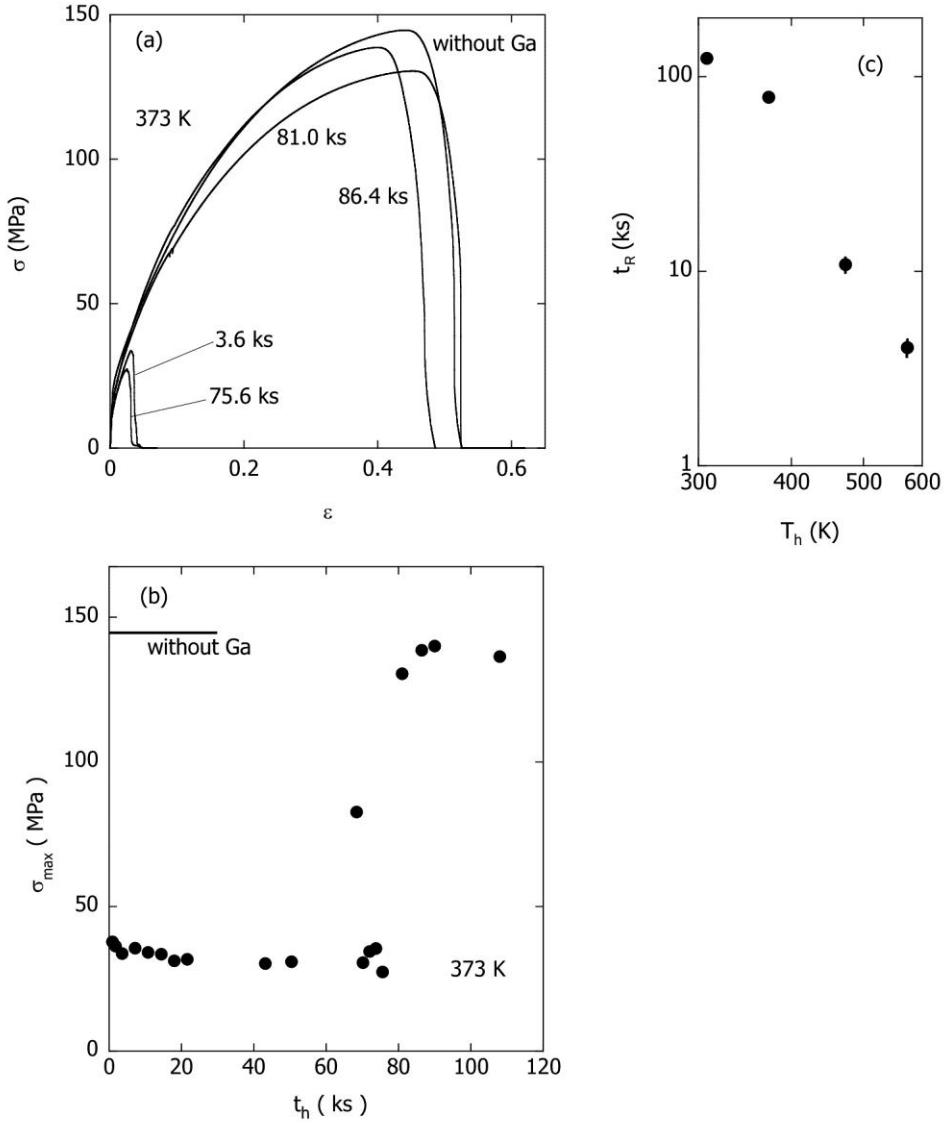

Figure 5. (a) The stress-strain curves. After the specimens were kept in contact with a small amount of gallium at 373 K for the time indicated in the figure, the tensile testing was performed at 308 K with a strain rate of 1.2×10^{-2} s^{-1}. (b) The maximum stress σ_{max} as a function of holding time t_h. (c) The time to recover ductility t_R as a function of the temperature of the furnace T_h [7].

is ductile despite gallium in liquid phase on the surface, and, therefore, the recovery of ductility at 465 K is not considered to be induced by formation of the intermetallic compound.

Various mechanisms of LME have been proposed, and it is still controversial whether crack initiation or crack propagation is a controlling process [1]. The specimen pulled at 423 K did not indicate clear necking (Fig. 4(c)), and the stress-strain curve shows abrupt drop after it reaches the maximum (Fig. 2). These indicate that a crack propagates very fast once a crack starts to move, and, therefore, a micro crack nucleation or start of crack motion seems to be especially blocked at this temperature.

5. Conclusion

Polycrystalline silver is embrittled by liquid gallium. Recovery from the embrittlement occurs if the contact time with liquid metal is long. This is considered induced by the formation of an intermetallic compound. Ductility trough is also observed as a function of test temperature in this solid and liquid metal couple. However, this ductility trough is not connected to forming the intermetallic compound.

References

[1] for a review: B. Joseph, M. Picat, F. Barbier, Liquid metal embrittlement: A state-of-the-art appraisal. Eur Phys J AP, 5 (1999) 19-31.
[2] F. A. Shunk, W. R. Warke, Specificity as an aspect of liquid metal embrittlement. Scripta metall, 8 (1974) 519-526.
[3] G. Nicaise, A. Legris, J. B. Vogt, J. Fost, Embrittlement of the martensitic steel 91 tested in liquid lead. J Nucl Mater 296 (2001) 256-264.
[4] K. Ina, H. Koizumi, Penetration of liquid metals into solid metals and liquid metal embrittlement. Mater Sci Engng A 387-389 (2004) 390-394.
[5] S. P. Lynch, Metal-induced embrittlement of materials. Mater Characterization 28 (1992) 279-289.
[6] K. Sakai, H. Koizumi, Time dependent embrittlement of polycrystalline Ag by liquid Ga, Procedia Engng 10 (2011) 643-648.
[7] K. Arakawa, K. Yamamoto, K. Sakai, H. Koizumi, to be submitted.