LASER-ULTRSONIC STUDY OF GALLIUM PENETRATION INTO ALUMINUM ALLOYS

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Abstract

We have studied penetration process of liquid gallium into 2017 aluminum alloys by measuring Rayleigh-wave velocity and attenuation using laser-generated ultrasound. The gallium penetration is the initial stage of liquid-metal embrittlement, which phenomenon is the reduction in the elongation to failure that can be produced when normally ductile solid metals are stressed while in contact with a liquid metal such as gallium. The results in unannealed samples showed that the Rayleigh-wave velocity decreased a few percent in the time scale of 10^3 s, suggesting a extraordinary-fast penetration of gallium along surface. The results in annealed samples showed the decrease in the time scale of 10^4 s. The difference of the penetration speed suggests that residual stress may facilitate the penetration of gallium.

Introduction

Aluminum and aluminum alloys, which are normally ductile solid metals, undergo liquid-metal embrittlement (LME) when attached with liquid gallium. This phenomenon has been studied extensively by tensile tests [1], however microscopic mechanism is unclear. Recently acoustic emission measurements [2] suggested the fast penetration of gallium into aluminum without tensile stress. Hugo and Hoagland [3] observed the gallium atoms penetration along grain boundaries of polycrystalline aluminum by means of a transmission electron microscope. Tanaka et al. [4] measured the penetration speeds of gallium into aluminum thin films to be 9 μm/s for 1000 nm -thick film.

The gallium penetration process may have the key to understand the liquid-metal embrittlement. The whole penetration process has not been analysed with bulk sample. The purpose of the present paper is to observe the penetration process by measuring ultrasonic velocity and attenuation with laser-generated technique. We utilized Rayleigh wave, longitudinal and shear waves for monitoring the process.

Experiments

A pulsed laser line source was used to excite ultrasonic Rayleigh waves in aluminum alloys. The experimental setup is shown in Fig.1. The excitation

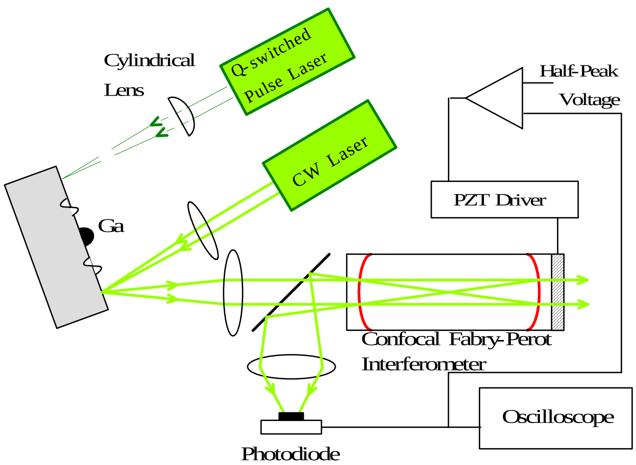

Figure 1 : The experimental setup.

laser was a Q-switched Nd:YAG laser (Continuum Minilite II, 532 nm) with the power of 35 mJ and the pulse duration of 5 ns. A single-frequency cw Nd:YAG laser (Lightwave 142H, 532 nm) with the power of 200 mW was used as a detecting probe. A confocal Fabry-Perot interferometer with the focal length of 500 mm was employed to detect ultrasonic movements of a surface. Reflected output from the interferometer was received by a photodiode (Thorlabs, 50 MHz), and recorded by an oscilloscope (Agilent 54616B, 2 GHz sampling). A stabilization system for compensating the drift of the laser-frequency and the interferometer was incorporated.

The sample used was 2017 aluminum alloys. The sample size was 50x50 mm and the thickness was 20 mm. The surface was polished emery papers and the last finish was given with 1 μm-alumina solution.

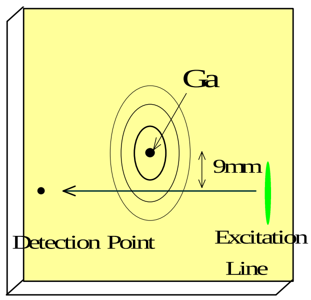

Figure 2 : The configuration of gallium deposition and the laser irradiation in the case of Rayleigh wave propagation..

The exciting laser beam was focused with a cylindrical lens to make a line source of 15 mm length. The distance between the source and the detection point is about 40 mm. The gallium deposition point was displaced 9 mm from the central axis of ultrasonic propagation, which configuration is illustrated in Fig.2. The gallium quantity used was 0.15g. The melting temperature of gallium is 29.8 °C, and so the sample temperature was kept at 35 °C by circulating thermostated water under the sample.

Longitudinal and shear waves measurements were also performed. The configuration for the longitudinal and shear waves measurements was different from that in Fig.1. The gallium was deposited to the bottom of a hall with 20 mm-depth and 4 mm-diameter, which drilled from the side. The laser irradiation points of excitation and detection were opposite to each other, so that the ultrasounds propagate just below the bottom of the hole.

In order to investigate the effect of residual stress the Rayleigh-wave measurement was carried out in annealed samples. The heat treatment was consisting the heating at 690 K for three hours, the cooling to 533 K with the rate of 1 K/min, and air quenching.

Results and Discussion

The time dependence of the Rayleigh-wave pulse was observed after gallium was put on the sample surface. The surface just under gallium was scratched with an edge to remove oxide films covering the surface. Gallium wetted to the surface and started to penetrate from this time. The waveforms were FFT analyzed to obtain spectra of amplitude and phase in a computer.

Gallium penetration in the sample without annealing

Figure 3 shows the time dependence of the amplitude of the Rayleigh-wave pulse relative to that with no gallium at several frequencies in the range of 4 - 10 MHz for the unannealed sample. The time dependence of the relative velocity obtained from FFT phase spectrum is shown in Fig.4 in the range of 2 -10 MHz. In Fig.3 two different processes are noticed: one is very rapid process within six minutes (to be referred to as the process A) and the other is slower process in the time range of 7 -26 min (to be referred to as the process B). On the other hand, the velocity change in Fig.4 suggests single process, which corresponds to the process B. The decrease in the amplitude initiated three minutes after the gallium deposition, while that in the velocity initiated five minutes after the gallium deposition. These results demonstrate that the process A is observed only in the attenuation measurement and not in the velocity measurement.

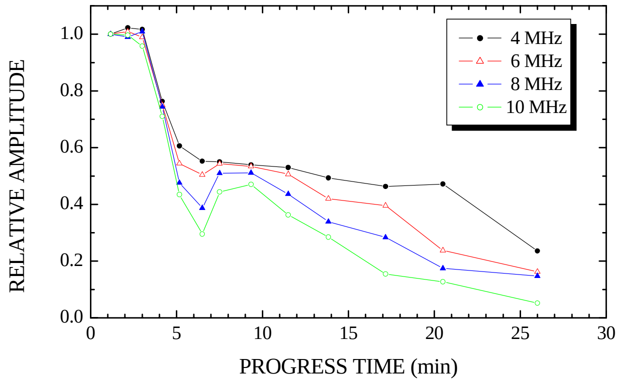

Figure 3 : Time dependence of relative amplitude of Rayleigh wave at several frequencies for Ga-deposited sample without annealing.

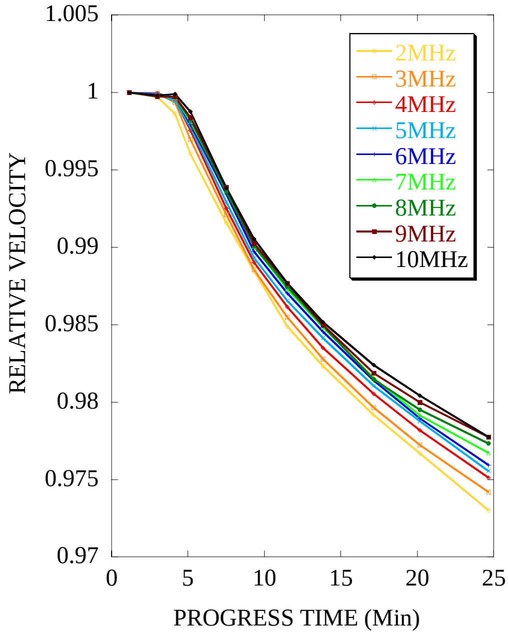

Figure 4 : Time dependence of relative velocity of Rayleigh waves for Ga-deposited sample without annealing.

The characteristic penetration speed might be estimated from the time when the amplitude decreased to a half value and the distance of 9 mm. The speed was obtained to be 30 μm/s for the process A, and 10 μm/s for the process B. These values are so extraordinary-large that the processes cannot be accounted by usual diffusion.

Figure 5 shows the time dependence of the relative velocities of longitudinal and shear waves Both velocities exhibited similar time dependence. Marked difference with the results for the Rayleigh wave is that the penetration speed into the bulk of the sample is two orders of magnitude slower than that for the Rayleigh wave. The speed is about 0.06 μm/s.

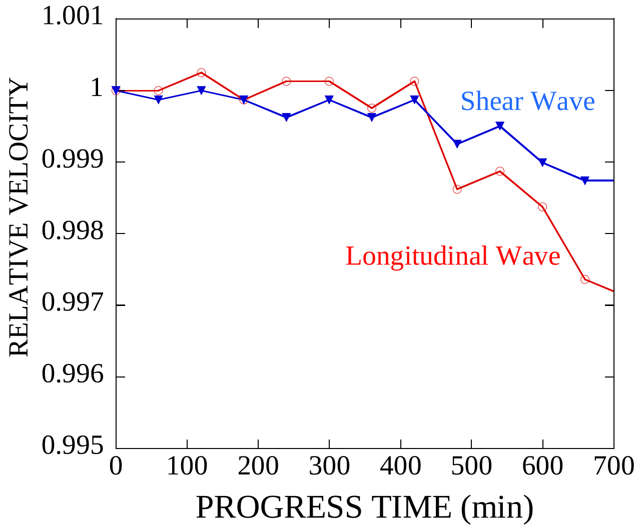

Figure 5: Time dependence of relative velocity of longitudinal and shear waves for Ga-deposited sample without annealing.

The gallium penetration was observed also in aluminum film evaporated on a glass substrate by Tanaka et al.[4]. The gallium placed on the aluminum film spreads like a spider's web at first with the speed of 2.3 – 9.1 μm/s depending on the film thickness and temperature. As the second stage, the continuous dull region extends with the speed of 0.8 μm/s, insensitively to the thickness and temperature.

Above results on the bulk and film samples suggest that the fast processes A and B are confined near surface and associated with the web-like penetration probably through grain boundaries. The distinction between the process A and B is not clear, but the process A is likely to associated with more straight penetration through surface with small quantity of gallium.

Effect of annealing on the penetration process

The time dependence of Rayleigh-wave pulse was observed for annealed sample until 540 min after gallium was deposited. Figure 6 indicates the relative amplitude after FFT analysis at 4, 6 and 8 MHz. The decrease in the amplitude is larger at higher frequencies. Figure 7 shows the time dependence of the relative velocity in the range of 3 – 9 MHz. In contrast with the results for the unannealed sample in Figs.3 and 4, the decrease in the amplitude and velocity occurs very slowly, about 20 times slower than in the unannealed sample. The speed was about 0.5 μm/s. The possible factors causing this difference are residual stress existing in the unannealed sample and growth of grain size by annealing. Unannealed sample has some residual stress that caused in manufacturing process. Based on the experimental observation that tensile stress facilitates embrittlement[5,6], residual stress may accelerate the gallium penetration. This indicates that the gallium

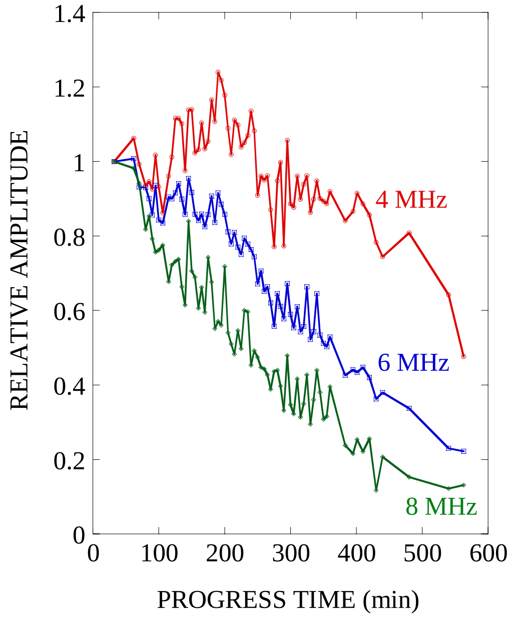

Figure 6: Time dependence of relative amplitude of Rayleigh wave for Ga-deposited annealed sample.

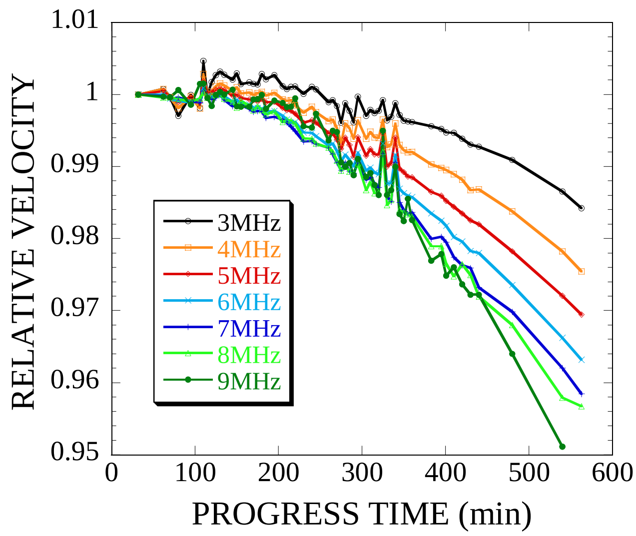

Figure 7: Time dependence of relative velocity of Rayleigh wave for Ga-deposited annealed sample.

penetration in unannealed sample is faster than in annealed sample which is free of the residual stress. Annealing also causes growth of grain size. The growth tends to reduce the penetration speed if the penetration proceeds through grain boundaries. Thus the annealing resulted in slowdown of the gallium penetration

Another difference is the inverse tendency of the frequency dependence of velocity. In Fig.4 the velocity decrease is larger at lower frequency, while in Fig.7 the velocity decrease is larger at higher frequency. Generally, displacement of Rayleigh wave is confined to superficial region of about one-wavelength depth. Then the displacement of higher-frequency Rayleigh wave is confined to more superficial region, then higher-frequency component is much more effected by gallium penetration. It is, therefore, reasonable that the velocity decrease is larger at higher frequencies, as in Fig.7 for the annealed sample. The inverse tendency for the unannealed sample in Fig.4 might be explained by the presence of the residual stress. If the degree of residual stress is greater in the interior than at the surface, the lower-frequency component is effected by the residual stress, resulting in the larger decrease in velocity at lower frequency.

Trace patterns of the gallium penetration

We can observe visually the trace of gallium penetration on the next day of the experiment. Figure 8 is the photograph of the unannealed sample surface (50 x 50 mm) taken 24 hours after the gallium deposition. The gallium was deposited at the center of the ellipse. Double ellipses exhibit the trace of gallium penetration into the bulk. The processes A and B in Figs.3 and 4 are not corresponding to the ellipses. The traces corresponding to the processes A and B are not visible and may exist outside the ellipses.

Figure 9 shows the photograph of the annealed sample surface (20 x 20 mm) taken 24 hours after the gallium deposition. The brightest circle in the center is the deposited gallium and outer circle represents the thick part of the penetrated gallium. The thin part extends outside the region of Fig.9. The trace pattern is circle in the annealed sample in contrast with the ellipse in the unannealed sample. The ellipse is probably associated with the directivity of the residual stress that remained in manufacturing process.

Figure 8: Photograph of the surface taken 24 hours after the Ga deposition for the sample without annealing. Double ellipses indicate the trace of Ga penetration.

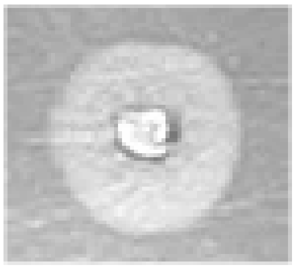

Figure 9: Photograph of the surface taken 24 hours after the Ga deposition for the sample with annealing. The brightest circle is the deposited Ga.

Conclusion

Gallium penetration processes into aluminium alloys were observed with laser-generated Rayleigh, longitudinal and shear waves. Two processes A and B were found in sample without annealing, and are associated with web-like penetrations along surface. Their speed was estimated to be 30 μm/s and 10μm/s, respectively. For annealed sample the speed of penetration along surface was found to decrease to be 0.5 μm/s. The difference was attributed to the release of residual stress and growth of grain size after annealing.

References

[1] M. G. Nicholas and C. F. Old, “Review Liquid metal embrittlement” J. Mater. Sci., vol.14, pp.1-18, 1979.

[2] Y. Kodama et al.,”Acoustic emission associated with the fracture of Al and Al alloy embrittled by liquid gallium” Mater. Sci. Engi., vol.A176, pp.231-235, 1994.

[3] R. C. Hugo and R. G. Hoagland, “In-situ TEM observation of aluminum embrittlement by liquid gallium” Scr. Mater. vol.38, pp.523-529, 1998.

[4] R. Tanaka, P.-K. Choi, H. Koizumi and S. Hyodo, “Fast penetration of liquid gallium in polycrystalline aluminum films” Mater. Trans. vol.42, pp.138-140, 2001.

[5] W. Rostoker, J. M. McCaughey and H. Markus, *Embrittlement of Liquid Metal*, Rheihold, N. Y.,1960.

[6] T. Mae and S. Hori,”The effect of stress on the liquid gallium embrittlement of aluminum”, Keikinnzoku, vol.37, pp.141-145,1987.