In Situ Observations of Solidification and Melting of Aluminum Alloy Using Ultrasonic Waveguide Sensor

Dikky Burhan*, Ikuo Ihara and Yoshihisa Seda*

Department of Mechanical Engineering, Nagaoka University of Technology, Nagaoka 940-2188, Japan

The in situ observations of solidification and melting of an aluminum-silicon alloy (Al–12.6%Si) using an ultrasonic waveguide sensor are presented. The ultrasonic sensor consists of a conventional piezoelectric ultrasonic transducer, a cooling system and a titanium (Ti) rod as an ultrasonic waveguide. The sustainability of the Ti rod in the molten Al alloy is investigated by immersion tests for 1, 4, 8 and 16 h at 800°C. The formation of a layer consisting of globular TiAl3, disperse AlSi2Ti and α-Al phases has been observed at the interface between the Ti and the Al alloy. Ultrasonic pulse-echo measurements of the Al alloy during solidification and melting have been performed using the ultrasonic sensor in temperature range from 200 to 800°C. The longitudinal wave velocity of the Al alloy shows a rapid and significant change from about 3900 to 5600 m/s around the eutectic point. An attempt to measure a solid/liquid interface of the Al alloy has been made at frequency of 2.25 MHz. The reflected echo from the interface undergoing directional solidification has been observed. The position and growth rate of the interface have also been determined from the reflected echo.

(Received May 9, 2005; Accepted August 2, 2005; Published September 15, 2005)

Keywords: in situ observation, aluminum–silicon alloy, ultrasonic measurement, titanium rod, solid/liquid interface, solidification

1. Introduction

The material properties of metals depend on microstructures produced during solidification. Since the solidified structure is difficult to eliminate once they are developed, it is important to control solidification process appropriately to prevent the unwanted structures. To make such process control, it is necessary to observe and understand solidification phenomena such as the kinetics of the formation of the solid/liquid interface. However, little technique is available for such in situ observation of solidification. It would be beneficial to have a technique that provides the in situ monitoring of solidification and melting.

Ultrasound, owing to its capability to probe the interior of materials, is a promising candidate for such in situ observation. Several ultrasonic techniques are being applied to high temperature measurements.1–20) The concept of an ultrasonic waveguide is a common technique for molten metal measurements.6–13) This technique is rather classical, but it is still attractive because of its simplicity, robustness and cost effectiveness. In this technique, measurements are made using a waveguide while the probe end is immersed in molten metals. Several attempts to observe molten metals using waveguides have been reported.6–13) However, they are not very successful because of the appearance of spurious echoes in measurements. Such spurious echoes are caused by the dispersion, wave reverberation, mode conversion and diffraction within the waveguide. The spurious echoes degrade the signal to noise ratio (SNR) because of their possible interference with the desired signals. In order to overcome such difficulty, recently clad waveguides with superior ultrasonic wave guidance and high SNR have been developed.14,15) Using such clad waveguides, several ultrasonic pulse-echo measurements have been successfully performed for molten Zn,16) Al17,18) and Mg19,20) at temperature up to 800°C.

*Graduate Student, Nagaoka University of Technology.

In this work, a new clad waveguide is used for in situ observation of Al alloy during solidification. Although a stainless steel was used as waveguide materials in the earlier works,16–20) it is certain that the steel waveguide is slowly and definitely attacked by chemical reactions with molten Al. We have chosen titanium (Ti) as the waveguide material because the sustainability of Ti in molten Al is higher than that of steel.21–23) In addition, it is expected from a good coupling of Ti with molten Al24) that ultrasonic energy can be effectively transmitted from the probe end of the Ti waveguide into molten Al so that pulse-echo measurements with high SNR can be made easily. The purpose of the present study is to demonstrate the practicability of a Ti waveguide for ultrasonic in situ observations of melting and solidification of an Al alloy. The sustainability of the Ti in the molten Al is evaluated and then ultrasonic pulse-echo measurements are performed in temperature range from 200 to 800°C.

2. Ultrasonic Waveguide Sensor

In this work, a Ti rod is used as an ultrasonic waveguide. The advantages of Ti waveguide are: (1) it is possible to transmit ultrasonic energy effectively from the waveguide into molten Al alloy because of high transmission coefficient between the two media, (2) it is expected that Ti has high temperature corrosion resistance to molten Al because of the formation of a stable and highly adherent protective layer on its surface.21–23) Table 1 shows the calculated reflection and transmission coefficients, R and T, for the interface between Ti and Al.

The interface between steel and Al. The R and T are determined from:

\[
R = \frac{\rho_2 c_2 - \rho_1 c_1}{\rho_1 c_1 + \rho_2 c_2} \\
T = \frac{2 \rho_2 c_2}{\rho_1 c_1 + \rho_2 c_2}
\]

where \(\rho\) and \(c\) are the density and the longitudinal wave velocity, respectively and subscript 1 and 2 denote the sensor
2108 D. Burhan, I. Ihara and Y. Seda

energy leakage from the side surface into molten Al. The tapered surface of the Ti rod in order to prevent ultrasonic spurious echoes. A tapered rod with tapering angle of 1.4° is used. A thermal sprayed Ti cladding is deposited on the tapered surface of the Ti rod in order to prevent ultrasonic energy leakage from the side surface into molten Al. Ultrasound can be effectively transmitted from Ti into Al and therefore, Ti is preferable for a waveguide material.

The basic construction of this sensor is almost the same as the ones used in the previous works, except for the waveguide material. The length and diameter of the Ti waveguide are 300 and 30 mm, respectively. In order to reduce the spurious echoes, a tapered rod with tapering angle of 1.4° is used. A thermal sprayed Ti cladding is deposited on the tapered surface of the Ti rod in order to prevent ultrasonic energy leakage from the side surface into molten Al. The cladding thickness is approximately 1.5 mm. The UT end of the waveguide is air-cooled by the cooling pipe to maintain the temperature below 50°C so that the conventional piezoelectric UT functions properly while the probe end of the waveguide is immersed in molten Al alloy at 800°C. The flow rate and pressure of the air cooling are 0.43 m³/min and 7 x 10⁴ kg/m², respectively.

3. The Sustainability Investigation

In order to perform molten Al alloy measurements, it is important to make sure that the Ti waveguide is not deteriorated during immersion for long time. For this purpose, immersion tests of Ti rods are conducted for 1, 4, 8 and 16 h in a molten Al alloy at 800°C. The same tests are done for stainless steel rods (JIS: SUS 410), for comparison.

The chemical compositions of the materials used are shown in Table 2. In the experiments, 2 kg of the Al alloy is melted in a crucible and Ti specimens are then immersed. The diameter and length of the specimens are 15 and 80 mm, respectively. Figure 2 shows the exteriors of the stainless steel and the Ti rods after immersion. The photograph clearly shows that the Ti rods can maintain their shapes even after 16 h of immersion, while the stainless steel rods are severely attacked by corrosion. Thus, the sustainability of Ti is quite better than that of stainless steel.

To investigate the chemical reactions between Ti and molten Al alloy, variations of the microstructures at the Ti/Al alloy interface after the immersion tests are examined. Figure 3 shows the microstructures of the cross section at the Ti/Al alloy interface. In 8 h and 16 h, the thickness of the intermediate layer is growing and the globular phases are observed in the intermediate layer. In order to investigate the qualitative chemical compositions and the phases in the intermediate layer, a scanning electron microscope (SEM) and an electron probe micro analyzer (EPMA) with wavelength dispersive spectrometer (WDS) are employed. Figure 4 shows the SEM and EPMA elemental images in the vicinity of the outer wall of Ti. It is found that the intermediate layer consists of globular Ti rich phases, dispersed Al–Si–Ti phases and Al rich phases. From the quantitative analysis, it is found that the globular Ti

material (steel or Ti) and the Al (solid or liquid), respectively. The densities of Ti, steel and solid Al alloy are 4500, 7880 and 2660 kg/m³, respectively. The density of liquid Al is assumed to be 2470 kg/m³. The velocities of Ti, steel, solid Al measured at 550°C and liquid Al measured at 600°C are about 5500, 5600, 5600 and 3900 m/s, respectively. It can be seen from Table 1 that the transmission coefficients of the Ti/Al interface are higher than those of the steel/Al interface for solid and molten conditions. This means that ultrasonic energy can be effectively transmitted from Ti into Al and therefore, Ti is preferable for a waveguide material.

The basic construction of this sensor is almost the same as the ones used in the previous works, except for the waveguide material. The length and diameter of the Ti waveguide are 300 and 30 mm, respectively. In order to reduce the spurious echoes, a tapered rod with tapering angle of 1.4° is used. A thermal sprayed Ti cladding is deposited on the tapered surface of the Ti rod in order to prevent ultrasonic energy leakage from the side surface into molten Al. The cladding thickness is approximately 1.5 mm. The UT end of the waveguide is air-cooled by the cooling pipe to maintain the temperature below 50°C so that the conventional piezoelectric UT functions properly while the probe end of the waveguide is immersed in molten Al alloy at 800°C. The flow rate and pressure of the air cooling are 0.43 m³/min and 7 x 10⁴ kg/m², respectively.

3. The Sustainability Investigation

In order to perform molten Al alloy measurements, it is important to make sure that the Ti waveguide is not deteriorated during immersion for long time. For this purpose, immersion tests of Ti rods are conducted for 1, 4, 8 and 16 h in a molten Al alloy at 800°C. The same tests are done for stainless steel rods (JIS: SUS 410), for comparison.

The chemical compositions of the materials used are shown in Table 2. In the experiments, 2 kg of the Al alloy is melted in a crucible and Ti specimens are then immersed. The diameter and length of the specimens are 15 and 80 mm, respectively. Figure 2 shows the exteriors of the stainless steel and the Ti rods after immersion. The photograph clearly shows that the Ti rods can maintain their shapes even after 16 h of immersion, while the stainless steel rods are severely attacked by corrosion. Thus, the sustainability of Ti is quite better than that of stainless steel.

To investigate the chemical reactions between Ti and molten Al alloy, variations of the microstructures at the Ti/Al alloy interface after the immersion tests are examined. Figure 3 shows the microstructures of the cross section at the Ti/Al alloy interface. In 8 h and 16 h, the thickness of the intermediate layer is growing and the globular phases are observed in the intermediate layer. In order to investigate the qualitative chemical compositions and the phases in the intermediate layer, a scanning electron microscope (SEM) and an electron probe micro analyzer (EPMA) with wavelength dispersive spectrometer (WDS) are employed. Figure 4 shows the SEM and EPMA elemental images in the vicinity of the outer wall of Ti. It is found that the intermediate layer consists of globular Ti rich phases, dispersed Al–Si–Ti phases and Al rich phases. From the quantitative analysis, it is found that the globular Ti
rich phases, the dispersed Al–Si–Ti phases and the Al rich phases are identified as TiAl₃, Al₅Si₂Ti and α-Al phases, respectively.

Since the solubility of Ti in Al is about 0.7 at%, the Al surrounding the Ti rod is readily saturated with Ti. The TiAl₃ layer and α-Al are then formed by peritectic reaction of the saturated Al:

\[
\text{Al}^{(\text{sat})} \rightarrow \alpha\text{-Al}^{(\text{sat})} + \text{TiAl}_3^{(s)} \tag{3}
\]

After the TiAl₃ layer is developed at the Ti surface, Ti element diffuses through the TiAl₃ layer. The Al surrounding the TiAl₃ layer is again saturated with Ti and then the reaction (3) is producing the globular TiAl₃ and α-Al phases. Reaction (3) explains why some α-Al are found together with the globular TiAl₃ phases as shown in Fig. 4.

Thus, the reaction of Ti in the molten Al alloy has been investigated. The formation of a diffusion controlled intermediate layer seems to inhibit the reaction of Ti with the molten Al alloy and therefore, the Ti waveguide might be useful for molten Al measurements.

4. Ultrasonic Measurements in Molten Al Alloy

Figure 5 shows the experimental setup for pulse-echo measurements in molten Al alloy. The measurement system consists of the ultrasonic waveguide, a personal computer (PC), a temperature logger card (National Instrument™ 4351), a high speed digitizer card (National Instrument™ 5112) and an ultrasonic pulser-receiver. The pulser-receiver is used to generate and receive ultrasonic signals at repetition rate of 1 kHz. Using the measurement system, both temperature and the ultrasonic signal data can be monitored at real time. A steel reflector (JIS: S45C) is assembled at the probe end of the sensor to make normal incident of the ultrasonic wave impinging at the surface of the reflector so that accurate pulse-echo measurements can be performed. The steel reflector is coated with zinc oxide (ZnO) to prevent a reaction with the melt. The sensor is immersed into the melt and pulse-echo measurements at frequency of 5 MHz are then performed.

Figure 6 shows the measured reflected echoes in the molten Al alloy at 800°C. R₁, R₂, R₃ denote the reflected echoes from the probe end, the surface and backside of the steel reflector, respectively. Immediately after the immersion, no significant echoes are observed except R₁ as shown in the top two traces in Fig. 6. There is almost no change in the echo even after 30 s as shown in the second trace. However, the amplitude of R₁ is reduced to about 44% after 50 s as shown in the bottom two traces. Simultaneously, the reflected echoes R₂ and R₃ have clearly appeared. This indicates that the ultrasonic coupling between the Ti rod and Al alloy occurs efficiently. The amplitude reduction is almost equal to the value of the reflection coefficient at the interface between the Ti and molten Al (approximately 44%) shown in Table 1.

4.1 Longitudinal wave velocity measurement

In order to investigate the variation in the longitudinal wave velocity at solid and molten states of the Al alloy, the melt is cooled down from 800°C to 200°C and then reheated. The cooling and heating rate are about 1 and 3°C/min, respectively. The velocity \( v \), is determined from:

\[
v = \frac{2L}{t} \tag{4}
\]

where \( L \) is the propagating distance of ultrasonic pulse wave and \( t \) is the transit time of the wave. For the velocity measurement of the Al alloy, the propagating distance and
transit time between the probe end and the reflector are used as shown in Fig. 6. The distance between the probe end and the reflector is corrected by taking into account the linear expansion of steel and Ti at elevated temperatures. The transit time is determined from echoes, $R_1$ and $R_2$, by the cross-correlation method.\(^{29)}\) Furthermore, to determine the temperature dependence of the velocities for the steel reflector and the Ti rod which are used for calculating the
reflection and transmission coefficients shown in Table 1, additional pulse-echo measurements are performed during heating and cooling. The velocity of the steel reflector is determined from $R_2$ to $R_3$. The velocity of the Ti rod is determined from $R_1$ and another echo at a step located at 20 mm from the probe end as shown in Fig. 6.

Figure 7(a) shows the variation in the longitudinal wave velocity for the Al alloy. It can be seen that in cooling process, the velocity of Al alloy significantly increases from about 3900 to 5600 m/s around the eutectic point at which solidification occurs. After the solidification, the velocity increases slightly as temperature decreases to 200°C. Similar velocity change can be found in heating process. It is noted that, a stepwise change in the velocity curves for cooling is observed around the eutectic point as shown by a circle in Fig. 7(a), which is not found in heating process. The velocity in the stepwise may arise from a mushy state where the solid and molten Al coexist at a given time during solidification. Although the detail of the mushy state is not known at this moment, it is reasonable to suppose that the velocity in the stepwise reflects the property of the mushy state whose velocity is intermediate between solid and molten states. Since the heating rate during melting (3°C/min) is higher than the cooling rate during solidification (1°C/min), no such stepwise is observed in heating process.

Figures 7(b) and (c) show temperature dependences in the longitudinal wave velocities for steel (S45C) and Ti, respectively. Both velocities decrease as temperature increases. A rapid change in the velocity of the steel is found at phase transformation points, $A_1$ or $A_3$, as shown in Fig. 7(b).

4.2 Solid/liquid interface measurements

An attempt to measure a solid/liquid interface of the Al alloy has been made. Figure 8 shows a schematic of the experimental setup used. In the experiment, 1 kg of ingot is melted in a directional solidification furnace and the Ti waveguide sensor shown in Fig. 1 is immersed into the melt at temperature of 700°C. The diameter and depth of the melt in the crucible are about 65 and 130 mm, respectively. Rotary and tilting stages are employed to make adjustment of the sensor. The same ultrasonic measurement system with PC as shown in Fig. 5 is used.

In the preliminary stage, a stable interface whose geometry is almost flat and location is fixed, is produced under an
appropriate temperature gradient using the directional solidification furnace. Ultrasonic pulse-echo measurements at frequency of 2.25 MHz are then performed to acquire the reflected echoes from the interface. Figure 9 shows the reflected echoes from the interface of Al alloy while the distance between the probe end and the interface is changed by moving the sensor position. The numbers in the Fig. 9 denote the distance in mm. The distance was determined from eq. (4) with two measured values: the measured velocity of molten Al shown in Fig. 7(a) and the transit time between the probe end echo and the interface echo shown in Fig. 9. Cross-correlation method is used for determining the transit time precisely.\(^{29}\) We can clearly see the echo from the stable interface and its shift as function of the distance. The amplitude of the echo is small because the reflection coefficient at the interface is about 0.21 that means that only 21.3% of the ultrasonic energy is reflected at the interface. Base on the preliminary result, monitoring of an interface during heating and cooling has been made. In the experiment, the distance from the probe end to the bottom of the crucible is fixed to be 45 mm. In the beginning of the experiment, the upper region of the furnace is heated to move the interface downward, and then the bottom of the crucible is rapidly cooled by spraying water at 5.2 ml/s under a pressure of 0.7 MPa to move the interface upward. Figure 10 shows the measured echoes from the moving interface as function of measurement time \(t\). Only 18 traces of the acquired signals are extracted at every 20 s increment and showed in Fig. 10. Since the observed interface echoes are tiny, shadowing is used to indicate the region of the liquid phase in Fig. 10. Although the interface echoes are tiny, we can recognize that the echo moves to the right hand during heating and then moves to the left hand after the water cooling. This movement of the echo corresponds to the behavior of the solidification front during melting and solidification.

Figure 11 shows the variations in the amplitude and the location of the echo from the solid/liquid interface during heating and cooling. The location \(L\) means the distance from the probe end and is determined from the transit time of the interface echo. By differentiating the location with respect to time, it is found that the moving speeds of the interface during heating and cooling are approximately 0.04 and 0.12 mm/s, respectively. We can also see a significant reduction in the amplitude of the probe end echo at about 300 s. This means that the solid/liquid interface is reached to the probe end at that time. Furthermore, we observe period-
In Situ Observations of Solidification and Melting of Aluminum Alloy Using Ultrasonic Waveguide Sensor

5. Conclusion

An ultrasonic waveguide sensor using a Ti rod has been prepared and applied for in situ observations of Al-Si alloy in temperature range from 200 to 800°C. The main results obtained are as follows:

1) Ti is preferable as a material of an ultrasonic waveguide sensor from the viewpoint of ultrasonic energy and sustainability in molten Al alloy. It is found that an intermediate layer consisting of globular TiAl3, dispersive AlSi2Ti and α-Al phases makes the reaction of Ti with the molten Al alloy inhibited.

2) Ultrasonic pulse-echo measurements are successfully performed at 5 MHz during heating and cooling and the longitudinal wave velocity of the Al alloy is monitored as a function of temperature. The velocity shows a rapid and significant change from about 3900 to 5600 m/s around the eutectic point.

3) An attempt to measure a solid/liquid interface of the Al alloy has been made at frequency of 2.25 MHz. The reflected echo from the interface undergoing directional solidification has been observed. From the observed echo, it is possible to determine the position and growth rate of the interface. The ultrasonic technique has the potential to be a useful diagnostic tool for in situ observation of solidification.

Acknowledgment

The authors are grateful to T. Okuno of Toyota Motor Co. for his support in the molten metal experiments, and Professor S. Kamado of Nagaoka University of Technology for his help on the material evaluation and analysis. Financial support from the Grant-In-Aid for Scientific Research (B14350397) by JSPS is appreciated.

REFERENCES