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Ultrasound assisted solidification process of ternary Cu–Sn–Sb alloy

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Abstract It is well-known that the application of ultrasound during liquid to solid transitions for alloys can refine the solidification microstructure and thus improves the mechanical properties. However, most published work focuses on single phase dendritic growth, whereas little has been conducted on the multiphase alloys with complicated phase transformations during solidification. In this work, the solidification process of ternary $Cu_{40}Sn_{45}Sb_{15}$ alloy was realized within intensive ultrasonic field with a resonant frequency of 20 kHz and ultrasound power from 0 W to 1000 W. The ultrasound refines the size of the primary ε(Cu_3Sn) intermetallic compound by two orders of magnitudes. If the ultrasound power increases to 1000 W, η(Cu_6Sn_5) phase nucleates and grows directly from parent liquid phase without the occurrence of peri-eutectic reaction on the top of the alloy sample where the ultrasound intensity is sufficiently high. These microstructural variations lead to the enhancement of compressive strength and elasticity modulus of ternary $Cu_{40}Sn_{45}Sb_{15}$ alloy.

1. Introduction

The microstructures of aerospace materials determine their applied properties. The solidification process of light alloys and superalloys for aerospace industries can be improved by ultrasound.[1–4] Thus the solidification of alloys within ultrasonic field has been an important research subject in recent years. The propagation of ultrasonic wave not only transmits acoustic energy into the liquid alloy, but also brings about such nonlinear effects as cavitation and acoustic streaming, which greatly affect the solidification process of alloys and ultimately improve their microstructures and mechanical properties.[5–8] So far, most work is focused on the dendritic growth of Al-based alloys[9–12] and Mg-based alloys[13–17] because of their wide applications in the automotive and aerospace industries.[18,19]

The peri-eutectic transition $L + \alpha \rightarrow \beta + \gamma$ is a typical kind of peritectic transformation in ternary and multicomponent alloys. Like the usual peritectic transition $L + \alpha \rightarrow \beta$, its reactants involve one liquid phase and one solid phase. However, its products are two cooperative solid phases. Since peri-eutectic transformation is an atomic diffusion controlled process which is difficult to be completed, the final microstructures are always composed of both primary and peri-eutectic phases.[20] Therefore, to control phase selection during peri-eutectic solidification is of great significance. However, the research of ultrasonic solidification of ternary

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peri-eutectic alloy is rather scarce. It can be speculated that if ultrasound is introduced into the solidification process of peri-eutectic alloy melt, the cavitation effect introduced by intensive ultrasound may alter the competitive nucleation between primary and peri-eutectic phases. Meanwhile, the acoustic streaming accelerates the solute transportation process, which may facilitate the peri-eutectic transformation.

Ternary Cu–Sn–Sb alloy is a complex metallic system whose solidification process involves multiple peri-eutectic transitions among various Cu–Sn, Sn–Sb and Cu–Sb intermetallic compounds. This makes it a good candidate of soldering materials without the toxic Pb element, because the formation of various intermetallic compounds could induce a significant composite strengthening effect, and thus has great potential applications in electronic interconnections of packing technology and automotive industry. In this work, the intense ultrasonic field is introduced into the solidification process of ternary $Cu_{40}Sn_{45}Sb_{15}$ alloy to investigate the microstructural evolution and mechanical property versus ultrasound power. On the basis of the experimental results, the peri-eutectic solidification mechanism is discussed.

2. Materials and method

The experiments were performed with a solidification apparatus incorporated with ultrasonic generator. The ternary $Cu_{40}Sn_{45}Sb_{15}$ alloy sample was $\varnothing$25 mm × 25 mm in size and was prepared from pure Cu (99.99%), Sn (99.99%) and Sb (99.99%) elements by using electrical resistance furnace. During the experiment, the alloy sample was melted by an electrical resistance furnace in the flowing argon atmosphere. The ultrasonic generator consists of two parts: a $KNbO_3$ piezoelectric transducer with a resonant frequency of 20 kHz and an ultrasonic horn with an end diameter of $\varnothing$22 mm. During experiment, the temperature was monitored by positioning a NiCr–NiSi thermocouple at the alloy sample top. When the alloy melting temperature dropped to 100 K above its liquidus temperature, the ultrasonic transducer was turned on and longitudinal ultrasonic wave was introduced from the top of the sample downward into the melted alloy until it solidified completely. Different exciting currents were input to the ultrasonic transducer. The corresponding ultrasound power P_W was estimated to be 250, 500, 1000 W. After experiments, the alloys samples were vertically sectioned, mounted, polished and etched. The phase constitution and microstructure of solidified alloy samples were analyzed by X-ray diffractometer (XRD) and scanning electron microscope (SEM).

The compression performance of ternary $Cu_{40}Sn_{45}Sb_{15}$ alloy was tested by CSS44100 universal electronic testing machine. Specimens in a size of $\varnothing$4.0 mm × 4.0 mm were cut from the central part of each alloy sample solidified with different ultrasound powers, and the loading speed of mechanical testing machine was set to be 0.3 mm/min downward. To ensure the accuracy of test results, the compression without samples was also conducted to make a baseline correction of machine-stiffness.

3. Results and discussion

3.1. Acoustic field distribution within liquid alloy

The sound distribution within liquid $Cu_{40}Sn_{45}Sb_{15}$ alloy is calculated by COMSOL Multi-physics 5.0 software. The propagation of ultrasound within liquid can be expressed by Helmholtz equation:

$$\frac{k^2}{\rho_0}p_a + \nabla\cdot\left(\frac{1}{\rho_0}\nabla p_a\right) = 0 \quad (1)$$

in which p_a is the sound pressure, $\rho_0 = 7746\ kg/m^3$ the density of ternary liquid $Cu_{40}Sn_{45}Sb_{15}$ alloy, $k = \omega/c_0$ the wave number, $\omega = 2\pi f$, $c_0 = 3750$ m/s the sound velocity, and $f = 20$ kHz the resonant frequency of ultrasound. The acoustic initial and boundary conditions are set as follows: (a) pressure source $p_a = p_0\cos(\omega t)$ at the ultrasonic probe tip surface and zero normal derivative of the pressure $\partial p_a/\partial n = 0$, where $p_0 = \sqrt{P_W\rho_0 c_0/S}$, t is the time, and $S = 3.14 \times 10^{-4}\ m^2$ is the surface area of the ultrasonic probe tip, (b) sound soft boundary $p_a = 0$ at the liquid/air interface, and (c) hard boundaries at the lateral and bottom sides of the crucible, which is written as:

$$-\frac{1}{\rho_0}(\nabla p_a) = 0 \quad (2)$$

Fig. 1(a) shows the sound distribution along the wave propagation direction z within the liquid alloy at $t = 0$ s. It can be seen clearly that the sound pressure increases with the increase

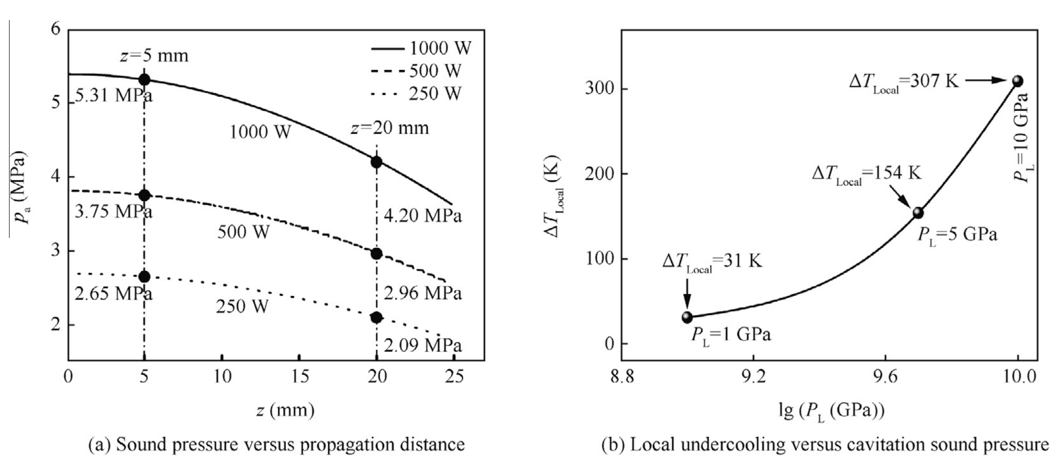

Fig. 1 Calculated sound pressure and local undercooling within liquid $Cu_{40}Sn_{45}Sb_{15}$ alloy.