Ultrasonic Infiltration in Alumina Fiber/Molten Aluminum System

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This study was aimed to improve the wettability of a non-wetting Al_2O_3 fiber/molten aluminum system, so as to attain full infiltration with a low applied pressure (P_a). The threshold infiltration pressure (P_c) of this composite system with an average fiber volume fraction (V_f) of 0.17 is 139 kPa at a molten aluminum temperature of 1023 K in pressure infiltration. Then non-infiltrated defects appear at locally high V_f portions. The θ of 2.24 rad is obtained taking into consideration the effect of non-infiltrated defects. Ultrasonic infiltration, in which ultrasonic vibration was transmitted into molten aluminum through a direct contact of the step-horn and melt, was performed to compare the apparent wettability with that in pressure infiltration. When molten aluminum was subjected to ultrasonic vibration with an ultrasonic power of 500 kW/m² (resonant frequency of 20.5 kHz), the P_c becomes zero, that is, the melt entry occurs even with a zero P_a . An infiltrated area with a semi-spherical shape is observed on the cross section close to the step-horn at the initial stage of ultrasonic infiltration, which coincides with the ultrasonic pressure (P_{us}) distribution obtained analytically. Then non-infiltrated defects are not observed in an incorporated region. Ultrasonic infiltration seems to have a kind of driving force giving aid to realize full infiltration, such as improvement of the apparent wettability due to a hysteresis of contact angles, the P_{us} acted instead of P_a and the high vibrational acceleration.

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I. Introduction

Although high pressure casting is one of the promising methods to fabricate metal matrix composites (MMC), the wettability between a reinforcement and molten metal strongly affects the characteristics and productivity of MMC. In wetting systems, full infiltration can be achieved without an applied pressure, and non-infiltrated defects are then inhibited from appearing at contact points of reinforcement. Unfortunately, most of the composite systems containing ceramic reinforcements are known to be non-wetted. The wettability of Al_2O_3 fiber⁽¹⁾⁽²⁾ and mica-ceramic particle⁽³⁾ by molten aluminum was estimated by a pressure infiltration method⁽¹⁾⁽⁴⁾, in which the reinforcement itself in MMC could be utilized. Then non-infiltrated defects appeared at contact points of reinforcement were clarified to affect the threshold infiltration pressure.

It is generally known that there are several methods to improve the wettability: addition of appropriate alloying elements to matrix metal⁽⁵⁾, surface treatment on reinforcements⁽⁶⁾⁽⁷⁾ and the optimum selection of processing parameters such as a molten metal temperature⁽²⁾. These are chemical approaches that make the contact angle lower through changing the balance of interfacial energies between molten metal, reinforcement and atmosphere. For example, the liquid processing accompanied by an exothermic reaction was investigated in a titanium powder premixed Al_2O_3 particle/molten

aluminum system, in which a molten aluminum temperature rose locally at the advancing molten metal front⁽⁸⁾. Then the threshold infiltration pressure became lower, and the volume of non-infiltrated defects reduced, because of the improved wettability.

Alternatively, the wettability can be also improved by a physical approach. One of the examples is an application of ultrasound to soldering: an aluminum/molten zinc alloy⁽⁹⁾ and an Al_2O_3 /molten zinc alloy/copper system⁽¹⁰⁾ have been soldered successfully, in which the wettability of molten zinc alloy was improved by ultrasonic vibration. A water droplet on the nylon substrate vibrated with 200–800 Hz was found to lower its contact angle compared with that on the stationary substrate⁽¹¹⁾, although it was not the case of ultrasound application.

In order to realize full infiltration with an extremely low applied pressure, the authors tried to improve the wettability by ultrasonic vibration as a physical approach. A water droplet on the paraffin coated substrate with ultrasonic vibration was found to lower the contact angle from a non-wetting to a wetting system. It was completely different from the contact angle determined by the equilibrium interfacial energies at a stationary state⁽¹²⁾. Pressureless infiltration was succeeded with assistance of ultrasonic vibration in an Al_2O_3 particle/molten aluminum system, which was selected as a model composite. Ultrasonic vibration also prevented non-infiltrated defects from their appearance at contact points of particles⁽¹³⁾.

The discontinuous Al_2O_3 fiber/molten aluminum system, which had been put into practical applications, was selected in this study. It was aimed to study the effect of ultrasonic vibration to molten aluminum on lowering the threshold infiltration pressure for the melt entry into a fiber preform, raising an infiltration rate and inhibitory action for non-infiltrated defect appearance.

II. Experimental Procedure

1. System of ultrasonic vibration

The ultrasonic vibration system consisted of a Langevin-type $\text{Pb}(\text{Zr}_x\text{Ti}_{1-x})\text{O}_3$ oscillator and a step-horn, as shown in Fig. 1. The step-horn made of titanium was utilized to gain a longitudinal vibration amplitude and to protect the oscillator from molten metal. Ultrasonic vibration was transmitted into molten metal through a direct contact of the step-horn and melt. TiB_2 was coated on the tip of step-horn by a spark coating method so as to avoid dissolution of titanium into molten metal.

The longitudinal amplitude of $2.5 \mu\text{m}$ at the oscillator end was amplified to $12.5 \mu\text{m}$ at the end surface of step-horn with an ultrasonic power (I_u) of 250 kW/m^2 in air atmosphere. The variation of longitudinal amplitude (b_u) at the end surface of step-horn as a function of I_u , which was measured by an optical displacement sensor, is shown in Fig. 2. The b_u increases rapidly with I_u , but it tends to saturate beyond I_u of 1000 kW/m^2 . When the step-horn tip is immersed in molten metal, the upward tendency of b_u is expected similar to that in air at-

Fig. 1 Schematic representation of PZT oscillator with a titanium step-horn.

Fig. 2 Variation of amplitude (b_u) measured by an optical displacement sensor as a function of ultrasonic power (I_u).

mosphere. However, the I_u dropped down a half of that in air atmosphere under the same current, since molten metal acted as a kind of load for vibration. Alternatively, the resonant frequency (f) of 20.65 kHz was measured by an impedance analyzer in air atmosphere. The f changed to 20.5 kHz in an immersed state, since the elasticity of titanium decreases with temperature. The I_u and f are the values measured in an immersed state, hereinafter, unless otherwise stated.

2. Preparation of fiber preform

Short $\delta\text{-Al}_2\text{O}_3$ fibers shown in Fig. 3 were supplied in this study. The average diameter (d_f) of $3.22 \mu\text{m}$ was obtained by a digitizer analysis of the scanning electron micrographs. Fiber preforms, which had a quasi-planar random fiber array, were prepared in the bottom of transparent quartz tubes with a closed end (inside diameter of 15.5 mm). A vibrating accumulation method for preform preparation was carried out in such a way that the fibers were disentangled with ultrasonic vibration in $0.05 \text{ mass\% SiO}_2$ sol aqueous solution and then naturally accumulated in the bottom of quartz tubes. Fiber preforms were fired at 1223 K for 7.2 ks in air atmosphere so as to improve the strength of preforms themselves. The height of preforms in quartz tubes was approximately 6.0 mm and the average volume fraction (V_f) of fiber preforms was 0.17 .

3. Ultrasonic infiltration

We designed the pressure infiltration apparatus, as shown in Fig. 4, which was consisted of a temperature and a pressure control system, an ultrasonic vibration system and a feedback circuit. Alternatively, commercially pure aluminum with $99.8 \text{ mass\%}$ purity was used as a matrix metal. An aluminum ingot of 8.0 g in mass was inserted on a fiber preform, then heated up in a low pressure of $(1-4) \times 10^{-3} \text{ Pa}$. Molten aluminum was maintained at a temperature of 1023 K for 300 s so as to

Fig. 3 Scanning electron micrograph of $\delta\text{-Al}_2\text{O}_3$ short fibers with an average diameter (d_f) of $3.22 \mu\text{m}$.

Fig. 4 Schematic representation of pressure infiltration apparatus with an ultrasonic vibration system.

homogenize a preform temperature. A molten aluminum surface was pressurized through an argon gas with a pressurizing rate of 0.33 kPa/s. The threshold infiltration pressure (P_c), which is the onset pressure of molten aluminum infiltration into a fiber preform during the pressurization, was measured by a visual observation of the interface between molten aluminum and fiber preform from the window opened at the side surface of heating furnace. After reaching the P_c , the pressure was successively risen up to a given applied pressure (P_a); the P_a was then maintained for a certain period (t_p).

The immersed end surface of step-horn was placed 1.0 mm above the molten aluminum-fiber preform interface. Ultrasonic vibration with I_u of 500 kW/m² was continuously transmitted from the beginning of pressurization to the termination of P_a retention. However, the P_a in ultrasonic infiltration was lower than that in pressure infiltration as was shorter the t_p . SiC fibers with $d_f=170$ μ m and an average length of 1.5 mm were also supplied as a fiber preform ($V_f=0.50$) to observe non-infiltrated defects easily. Fiber preforms incorporated with molten aluminum were taken out after their solidification, each non-infiltrated area was measured by a digitizer, and non-infiltrated defects appeared in the incorporated region were microscopically observed on the vertical cross sections.

III. Results and Discussion

1. Non-infiltrated area and non-infiltrated defects

A non-infiltrated area is defined as the area that molten aluminum does not incorporated with a fiber preform on the vertical cross section, and a non-infiltrated area ratio is a ratio of non-infiltrated area to a whole cross sectional area of fiber preform. Molten aluminum can penetrate into a fiber preform with an applied pressure (P_a) which is higher than the threshold infiltration pressure (P_c). If retention time (t_p) of P_a is too short, a non-infiltrated area remains resulting from a termination of the advancing molten aluminum front. Even if the t_p is long enough, the molten aluminum front may stop advancing at the distance of P_a balancing with the interfacial energy changes. Then non-infiltrated defects appear at contact

Fig. 5 Optical micrographs showing (a) infiltrated area and (b) non-infiltrated defects in the Al₂O₃ fiber/Al system pressurized with $P_a=180$ kPa for $t_p=300$ s.

points of fibers or locally high V_f portions.

A typical infiltrated area and non-infiltrated defects appeared at the top region of preform are shown in Fig. 5, which are taken on the cross section of the specimen prepared with $P_a=180$ kPa for $t_p=300$ s in pressure infiltration. A non-infiltrated area ratio is 0.40 in Fig. 5(a). Although non-infiltrated defects can be clearly observed at contact points of thick fibers or large particles which are non-wetted with molten aluminum⁽⁸⁾, in the case of thin fibers of Fig. 5(b), non-infiltrated defects are observed as a small void, in which molten aluminum cannot penetrate due to the irregularity of V_f .

2. Lowering non-infiltrated area and threshold infiltration pressure by ultrasonic vibration

Pressure infiltration with $P_a=180$ kPa for $t_p=300$ s cannot attain full infiltration of a whole fiber preform, as mentioned before. Typical optical micrographs of Fig. 6 show infiltrated areas in ultrasonic infiltrations with an ultrasonic power (I_u) of 500 kW/m², where Fig. 6(a) is the cross section of the specimen prepared with $P_a=0$ kPa for $t_p=10$ s and (b) with $P_a=140$ kPa for $t_p=10$ s. Molten aluminum can penetrate radially into a fiber

Fig. 6 Optical micrographs showing infiltrated area in ultrasonic infiltration of the Al₂O₃ fiber/Al system with (a) $P_a=0$ kPa for $t_p=10$ s and (b) $P_a=140$ kPa for $t_p=10$ s.

Fig. 7 Variation of non-infiltrated area as a function of applied pressure (P_a), threshold infiltration pressure (P_c) in pressure and ultrasonic infiltration of the Al_2O_3 fiber/Al system.

preform even with a zero P_a . However, the molten aluminum front stops advancing when a driving force of ultrasonic infiltration balances with the interfacial energy changes. Thus full infiltration cannot be attained for $t_p=10$ s with a zero P_a in ultrasonic infiltration. The infiltrated area of Fig. 6(a) with a semi-sphere is a characteristic shape of ultrasonic infiltration before reaching full infiltration. The full infiltration of Fig. 6(b) is attained by ultrasonic vibration with $P_a=140$ kPa for $t_p=10$ s, which is only one-thirtieth of the t_p in pressure infiltration. Hence, it is expected that an infiltration rate becomes higher due to ultrasonic vibration comparing with that in pressure infiltration.

The variations of a non-infiltrated area ratio are shown in Fig. 7 as a function of P_a in pressure and ultrasonic infiltration, respectively. Where the t_p is 300 s in pressure infiltration and 10 s in ultrasonic infiltration. The P_c in pressure and ultrasonic infiltration are also shown in Fig. 7. The P_c of 139 kPa in pressure infiltration, at which molten aluminum starts penetrating into an Al_2O_3 fiber preform, reduces to zero in ultrasonic infiltration. It is to be expected that molten aluminum can penetrate into a fiber preform with a zero P_a right after the initiation of ultrasonic vibration, in spite of the non-wetting system. If the P_a is higher than the P_c , a non-infiltrated area ratio rapidly decreases with increasing P_a . Full infiltration can be achieved by applying $P_a \geq 210$ kPa for $t_p=300$ s in pressure infiltration, and $P_a \geq 140$ kPa for $t_p=10$ s in ultrasonic infiltration. As mentioned previously, an infiltration rate is accelerated at least 30 times by ultrasonic vibration, so that full infiltration can be achieved within an extremely short t_p . The infiltration rate tends to increase qualitatively with P_a increased, although it cannot be measured quantitatively by a visual observation of the interface from the side window. The molten aluminum front proceeds during a retention period of P_a , a non-infiltrated area ratio then decreases with t_p lengthened at a certain P_a ($\geq P_c$).

3. Inhibitory action for non-infiltrated defect appearance

Non-infiltrated defects can be observed on the cross section of the top preform region infiltrated even with $P_a=210$ kPa for $t_p=300$ s in pressure infiltration, as shown in Fig. 8, although this pressurizing condition could realize full infiltration. Molten aluminum cannot penetrate into high V_f portions, which remain as a typical non-infiltrated defect in a thin Al_2O_3 fiber/molten aluminum system, in place of defects appeared at contact points of thick reinforcement.

In order to observe the other type of non-infiltrated defects, which may appear at the contact points of thick reinforcement, ultrasonic infiltration of molten aluminum was performed with $P_a=20$ kPa for $t_p=10$ s using a SiC fiber preform with $d_f=170$ μm and $V_f=0.50$. Its d_f is approximately 50 times thicker than that of the Al_2O_3 fibers. The comparison of non-infiltrated defects appeared in ultrasonic infiltration and pressure infiltration with $P_a=50$ kPa for $t_p=300$ s was carried out on scanning electron micrographs of Fig. 9. The P_c of 19.1 kPa is measured in pressure infiltration of a SiC fiber/molten aluminum system, the P_c also becomes zero in ultrasonic infiltration. In the case of pressure infiltration, non-infiltrated defects with a meniscus end generate at the contact points of the fibers or close to there. However, they entirely disappear in ultrasonic infiltration. Thus, ultrasonic vibration of molten aluminum has a favorable inhibitory action for non-infiltrated defect appearance.

A typical optical micrograph taken on the cross section of the infiltrated region, in which full infiltration has been achieved ultrasonically with $P_a=140$ kPa for $t_p=10$ s, is shown in Fig. 10. Non-infiltrated defects are not observed in any regions, although pressure infiltration has made non-infiltrated defects appear at locally high V_f portions even with a higher P_a than the P_c and a longer t_p than that in ultrasonic infiltration. The molten aluminum front with ultrasonic vibration apparently becomes a wetting system with Al_2O_3 fibers, because it shows a

Fig. 8 Optical micrograph showing non-infiltrated defect in Al_2O_3 fiber/Al system pressurized with $P_a=210$ kPa for $t_p=300$ s.

Fig. 9 Scanning electron micrographs showing non-infiltrated defect in the SiC fiber/Al system, (a) pressure infiltration with $P_a=50$ kPa for $t_p=300$ s and (b) ultrasonic infiltration with $P_a=20$ kPa for $t_p=10$ s.

Fig. 10 Optical micrograph showing full infiltration without non-infiltrated defects in ultrasonic infiltration of the Al_2O_3 fiber/Al system with $P_a=140$ kPa for $t_p=10$ s.

hysteresis between an advancing and a receding contact angle. This seems to be one of the reasons why ultrasonic vibration inhibits non-infiltrated defect appearance in a fiber preform.

If whole fiber surfaces are wetted thoroughly during the infiltration, the relation between the P_c and contact angle (θ) is expressed by eq. (1)⁽¹⁾.

$$P_c = -\{4\gamma_{la} \cos \theta / d_f\} \{V_f / (1 - V_f)\} \quad (1)$$

where d_f is the average fiber diameter, V_f the fiber volume fraction and γ_{la} the interfacial energy between molten aluminum and atmosphere.

With only one exception of perpendicular infiltration to an aligned fiber preform, the P_c does not depend on a fiber alignment. However, the fiber alignment strongly affects the volume of non-infiltrated defects appeared in a non-wetting fiber/molten metal system. Supposing a

non-infiltrated defect to appear at a contact point of two fibers with a meniscus end, the relation between P_c and θ is modified as follows⁽¹⁾.

$$P_c = \gamma_{la} [2X_{la} - \{2\pi d_f a (1 + \tan^2 \alpha + \tan^2 \beta)^{1/2} - 2X_{sa}\} \times \cos \theta] / [a^3 \tan \alpha \tan \beta - (1/2)\pi d_f^2 a (1 + \tan^2 \alpha + \tan^2 \beta)^{1/2} - 2X_v] \quad (2)$$

where X_v is the volume of non-infiltrated defect, X_{sa} the interfacial area of fiber/defect and X_{la} the interfacial area of molten aluminum/defect, and a standard side length (a) of the fiber array model is expressed in eq. (3)⁽¹⁾.

$$a^2 = \pi d_f^2 (1 + \tan^2 \alpha + \tan^2 \beta)^{1/2} / (2V_f \tan \alpha \tan \beta) \quad (3)$$

where α and β are the average horizontal and vertical alignment angle of fibers. Since the fiber alignment seems to be quasi-planar random in the Al_2O_3 fiber preforms resulting from a natural accumulation, α and β are then deduced to be $\pi/4$ rad and $\pi/18$ rad, respectively.

After the analytical calculation of X_v , X_{sa} and X_{la} , the θ of 2.24 rad is obtained through substituting $P_c=139$ kPa, $V_f=0.17$ and $\gamma_{la}=0.833 \text{ Pa}\cdot\text{m}^{(14)}$ into eq. (2). However, any non-infiltrated defects do not appear in ultrasonic infiltration, so that the P_c of 140 kPa is calculated through substituting $\theta=2.24$ rad into eq. (1). There seems to be a kind of driving force for infiltration of molten aluminum with ultrasonic vibration into a fiber preform. The driving force of ultrasonic infiltration becomes at least 140 kPa acted instead of P_a .

Hence, ultrasonic infiltration is expected to be a liquid processing for MMC productions with an extremely low P_a in the non-wetting composite systems, and ultrasonic vibration not only inhibits the appearance of non-infiltrated defects in an incorporated region, but also gains a rather fast infiltration rate.

4. Driving forces in ultrasonic infiltration

Strong non-linear effects appear adjacent to a sound source element in liquid acoustic field⁽¹⁵⁾. In order to simplify a distribution analysis of ultrasonic pressure (P_{us}), it is postulated that there are no reflections of ultrasound at the interfaces of quartz tube/molten aluminum and atmosphere/molten aluminum. The wave length of 224 mm is calculated through substituting the values of $f=20.5$ kHz and the sound velocity in molten aluminum of 4600 m/s at 1023 K⁽¹⁶⁾. Then it is also possible to ignore an effect of fiber existence, since the wave length of ultrasound is large enough comparing with d_f of the Al_2O_3 fibers. The acoustic field is supposed to be consisted of superimposed spherical waves from an aggregate of sound source elements using the polar coordinate system, as shown in Fig. 11.

The P_{us} at an observe point aparted by D from a step-horn center (O) is expressed in eq. (4)⁽¹⁷⁾:

$$P_{us} = \{j\omega\rho V_0 / (2\pi)\} \int_0^{2\pi} \int_0^r \{ \exp(-jk d_1) / d_1 \} \times d_2 dd_2 d\beta \quad (4)$$

where ω is the angular velocity, k the wave length coefficient, ρ the density, r the end surface radius of step-horn and V_0 is the effective vibration velocity at the end surface of step-horn, which is expressed in eq. (5).

$$V_0 = \omega b_u / 2^{1/2}. \quad (5)$$

According to eq. (5), the V_0 of 1.1 m/s is obtained. Then a distance from a sound source element to an observe point is expressed by eq. (6).

$$d_1 = (D^2 + d_2^2 - 2Dd_2 \sin \alpha \sin \beta)^{1/2}. \quad (6)$$

The curved plane of P_{us} distribution ahead of the end surface of step-horn is shown in Fig. 12, which is calculated by an engineering workstation through substituting $\rho=2400$ kg/m³ and $r=3$ mm into eq. (4).

Fig. 11 Polar coordinate system for the estimation of ultrasonic pressure.

Fig. 12 Computer drawing of ultrasonic pressure (P_{us}) distribution ahead of the step-horn due to superimposed spherical waves.

The equi-pressure plot on the P_{us} plane seems to be a quasi-semi-circle round the end surface of step-horn, which coincides with the infiltrated area shown in Fig. 6(a); ultrasonic infiltration proceeds radially from the center of top preform surface. Hence, infiltration occurs due to the high P_{us} in molten aluminum at the initial stage, but the P_{us} tends to decrease with infiltration advanced. This is the reason why the semi-spherical infiltrated area coincides with the above linear analytical equi-pressure plot.

In addition, the effective acceleration (α_{us}) is expressed in eq. (7):

$$\alpha_{us} = \omega^2 b_u / 2^{1/2}. \quad (7)$$

The α_{us} of 140 km/s² is obtained through eq. (7) in molten aluminum close to the end surface of step-horn, the α_{us} reaches 14000 times larger than the acceleration due to gravity. It is expected that the infiltration rate of molten aluminum is rather high in the vicinity of step-horn end, but it reduces with a distance between the molten aluminum front and step-horn aparted, in good agreement with the experimental results.

IV. Conclusions

In order to realize full infiltration of a short Al_2O_3 fiber/molten aluminum system with a low applied pressure (P_a), ultrasonic infiltration (resonant frequency of 20.5 kHz) was performed at 1023 K using Al_2O_3 fiber preforms with a fiber volume fraction (V_f) of 0.17. The following results were obtained:

(1) The threshold infiltration pressure (P_c) of 139 kPa is obtained in pressure infiltration, but it reduced to zero in ultrasonic infiltration, that is, molten aluminum starts into penetration without P_a .

(2) Since pressureless infiltration occurs due to improvement of the apparent wettability, full infiltration is achieved with a low P_a and within a short processing time in ultrasonic infiltration.

(3) Although non-infiltrated defects appear at locally high V_f portions in pressure infiltration, ultrasonic vibration of molten aluminum inhibits their appearance in an incorporated region.

(4) The contact angle (θ) of 2.24 rad is calculated through $P_c = 139$ kPa taking into consideration the effect of non-infiltrated defects. Since the P_c becomes zero and there appear no non-infiltrated defects in ultrasonic infiltration, its driving force acted instead of P_a is estimated at least 140 kPa.

(5) An infiltrated area with a semi-spherical shape is observed close to the step-horn at the initial stage of ultrasonic infiltration; it is coincided with the analytically obtained plane of ultrasonic pressure (P_{us}) distribution.

(6) Full infiltration with a low P_a is achieved in ultrasonic infiltration within a short processing time, because of improvement of the apparent wettability, the P_{us} acted instead of P_a and the extremely high vibrational acceleration.

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