

Sonoprocessing of wetting of SiC by liquid Al: A thermodynamic and kinetic study

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ABSTRACT

The sonoprocessing of droplet spreading during the wetting process of molten aluminum droplets on SiC ceramic substrates at 700 °C is investigated in this paper. When wetting is assisted by a 20 kHz frequency ultrasonic field, the wettability of liquid metal gets enhanced, which has been determined by the variations in thermodynamic energy and wetting kinetics. Wetting kinetic characteristics are divided into two stages according to pinning and depinning states of substrate/droplet contact lines. The droplet is static when the contact line is pinning, while it is forced to move when the contact line is depinning. When analyzing the pinning stage, high-speed photography reveals the evidence of oxide films being rapidly crushed outside the aluminum droplet. In this work, atomic models of spherical Al core being wrapped by alumina shell are tentatively built, whose dioxide microstructures are being transformed from face-centered cubic into liquid at the atomic scale. At the same time, the wetting experiment reveals that the oxide films show changes in the period of sonoprocessing from 3rd to 5th second.

During the ultrasonic spreading behavior in the late stage, there is a trend of evident expansion of the base contact area. The entire ultrasonic process lasts for no longer than 10 s. With the aid of ultrasonic sinusoidal waves, the wettability of metal Al gets a rapid improvement. Both molecular dynamic (MD) investigations and the experiments results reveal that the precursor film phenomenon is never found unless wetting is assisted by ultrasonic treatments. However, the precursor film appears near the triple line after using ultrasonics in the droplet wetting process, whose formation is driven by ultrasonic oscillations. Due to the precursor film, the ultrasonic wetting contact angle is lower than the non-ultrasonic contact angle. In addition, the time-variant effective ultrasonic energy has been quantitatively evaluated. The numerical expressions of thermodynamic variables are well verified by former ultrasonic spreading test results, which altogether provide an intrinsic explanation of the fast-decreasing contact angle of Al/SiC.

1. Introduction

Sonoprocessing has long been widely used in industrial applications that include ultrasonic degradation, ultrasonic catalytic synthesis, ultrasonic extraction, and ultrasonic preparations of nanomaterials [1–5]. In recent years, the application of ultrasonic treatment in liquid metal processing has increasingly gained attention. Such applications mainly focus on ultrasonic soldering and ultrasonic preparation of ceramic-reinforced metal-based composite materials [6–8]. Successful application of these techniques is determined by the wettability and spreading properties of liquid metals. However, the wettability of liquid metals and solid ceramics is relatively poor [9]. With the assistance of

ultrasonics, wettability can be improved. Both the sonoprocessing technique for wetting and the ultrasonic wetting mechanism have become common issues and key problems to be solved.

According to some existing research on the above-mentioned aspects [10–12], ultrasonic treatment greatly enhances the wettability of liquid metals on solid ceramics. Such metals include tin, zinc, and aluminum. The wetting processes of SiC by Al droplets are of great importance. First of all, joining metals with the SiC ceramic is desirable for a wide range of aerospace, automobile, electronic, and biomaterial sectors, thus, soldering of SiC at relative low temperatures is in demand in engineering field. The second, SiC-reinforced aluminum metal matrix composites are also fundamental functional and engineering materials with qualified

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mechanical properties [13–15]. Methods for improving wettability have already been investigated through experimental studies [16], however, the ultrasonic-assisted method has its unique advantage.

For liquid metals with poor wettability, sonoprocessing changes their flow behaviors and increases the systematic energy used for spreading [17–19]. For other liquid metals such as Al–10Cu or Bi–8Zn, they can spontaneously spread on solid substrates [20,21], but the viscosity relates with the wetting tendency and it is unique for each liquid alloy. The viscous force is significantly changed when assisted by the ultrasonic shock wave, and the ultrasonic treatment provides an extra driving force for moving liquids [22]. Xu et al. [23] have focused on the dynamic behavior of cavitation bubbles in molten metal alloys using X-ray imaging technology. They found that tiny bubbles that have emerged due to the high-power ultrasonic wave will shrink and then collapse within a life cycle of several acoustic periods.

Sonoprocessing greatly enhances the wettability of liquids inside reactive wetting systems. The reason behind wettability enhancement lies in the big difference between the metal alloy containing metal/metal bonds and the ceramic ionic bonds. Since the wettability between metal and ceramic is poor, the bonding strength between metal and ceramic is also weak. The mechanical properties of ceramic-reinforced metal-based composites also deteriorate, which leads to the unsatisfied performance of the respective material. Laurent et al. have previously studied the wettability of Al/SiC in a wide temperature range from 900 to 1100 °C [24,25]. They found that the oxidation of Al droplet surface was inevitable under vacuum in normal conditions, and under such circumstance, the contact angle was difficult to lower unless the temperature for wetting was kept rising. Al/SiC wetting with no ultrasonic induction has also been investigated by Bao [26], where the dynamic contact angle was estimated to revert from the initial obtuse angle to an acute angle at low speed. During the study of Bao, the time required for maintaining the high temperature for Al/SiC was longer than 50 min. Some scholars still hold the view that only with a high temperature accompanied by an extremely lower oxygen-pressure, can the wettability of Al on SiC receive a minor enhancement. If the chemical reaction between the Al droplet and SiC substrate activates, the reactant appears on the liquid/solid interface which is also good for wettability enhancement without ultrasonic assistance [27–29]. If the time of Al/SiC exposure to high temperature increases, the interface bond formed is of poor quality due to excessive brittleness of interfacial reaction products. Those interfacial reaction products weaken the joining strength of Al/SiC excessively.

By the aid of exerting power ultrasonics to wetting, the external acoustic energy is input into Al/SiC wetting system instead of so much heat [30], the holding temperature for this wetting system could decrease and the joining strength of Al/SiC pieces would be hardly weakened. So the unique advantage of the ultrasonic-assisted method greatly benefits the enhancement of the Al/SiC wettability.

The entire ultrasonic process ends within a short time period; therefore, the wetting process is not exposed to high temperatures for a long time and the brittle interfacial reactants can be avoided [31]. Ultrasonics can also impact the oxide layers, which can be broken to allow the fresh metal to come into contact with the solid in order to improve the wettability. Till now, relevant research has seldom been reported; hence, the inherent mechanism of liquid wettability by ultrasonic treatments is of great importance. On the one hand, the effect of ultrasonics on the instantaneous breakage behavior of oxide film on Al liquid is good for the wettability of Al/SiC, the process of which is generally known as cavitation [32,33]. Cavitation produces strong fluctuations in the liquid metal. On the other hand, sonoprocessing has been successfully used to remove oxide films on surfaces of aluminum alloys; however, neither the oxide removal mechanism of Al liquid nor the detailed kinetic law of ultrasonic wetting is clear.

In this paper, molecular dynamic simulations (MD) have been performed in order to visually analyze the process of oxide film breakage, wetting, and spreading under ultrasonics at the atomic scale. The phenomenon of precursor film is a key factor in the improvement of liquid

wettability. The precursor film structure during the ultrasonic spreading process effectively indicates that the wetting degree is improving. E. B. Webb et al. [34] have made the earliest attempt to use MD for the atomic wetting simulation with a Pb/Cu metal/metal system. According to their research, the rate of dissolution between metal alloys determines the flowing velocity of liquids during wetting. However, MD calculations of the metal/ceramic ultrasonic wetting have not yet been reported. Therefore, the breaking process of oxide films and the dynamic wetting behaviors after oxide removal, as well as the appearance of the precursor film structure, are visualized and analyzed via MD simulations in this work.

2. Preparation of experiments

2.1. Specimen materials and equipment

Metal Al is commonly used in high-temperature vacuum soldering. As a good filler material, Al has both good fluidity and a low melting point. Once there is no aluminum oxide, the high-temperature liquid can wet most of the ceramics. Table 1 lists the property parameters of both the Al solder piece and the ceramic substrate used in experiments.

Prior to the spreading test, clean ceramic substrates ultrasonically in acetone for 10 min. After drying, the SiC substrate sample is placed in the furnace, pinning on the inner platform with a homocentric squares mold. Then one piece of Al is placed on the substrate (after both ultrasonic and caustic wash) in the assembled furnace. Wetting occurs with the assistance of ultrasonic vibrations, as shown in Fig. 1. The furnace pressure is limited to 5×10^{-3} Pa with a molecular pump. Then the Al piece and SiC substrate are heated together. The heating rate is kept constant to a value close to 10 °C/min. A thermocouple is used to control the heating progress until the furnace temperature reaches 700 °C.

In this experiment, the power ultrasonic used in sonoprocessing is selected as 20 kHz, the value 20 kHz is the cusp of ultrasonic frequency. At high temperature, the ultrasonic wave propagates from the SiC substrate to Al, the internal friction is the main consumption of energy at the liquid/solid interface, thus it is equivalent to having the total ultrasonic output energy reduced. The ultrasonic internal friction is related to the frequency, the larger the frequency is the more the internal friction consumes, therefore the frequency is set as 20 kHz, so as to release the heat energy consumed by the internal friction, and then promote the wettability to the greatest extent. Ultrasound waves with higher frequency, 30 kHz or 40 kHz, result in larger degrees of internal friction.

The entire process is synchronously recorded by a CCD camera. Images are stored and then used for further post-processing. Videos and images are analyzed with the aid of Photo Line V17.54.

2.2. Details of molecular dynamics

2.2.1. A potential function for the oxide film removal simulation

The third-generation charge-optimized many-body (COMB3) potential function has been modeled for calculations of Al and O atoms [35]. According to COMB3, the N -atoms' potential energy is obtained by integrating Eq. (1), as follows:

$$E_i^{\text{tot}}(r, q) = E^{\text{self}}(r, q) + E^{\text{short}}(r, q) + E^{\text{Coul}}(r, q) + E^{\text{polar}}(r, q) + E^{\text{vdW}}(r) + E^{\text{corr}}(r) \quad (1)$$

Table 1
Features of the specimen materials.

Properties	Treatment processes	Bulk density, (g·cm ⁻³)	Acoustic velocity (m·s ⁻¹)	Melting temperature (°C)
aluminum (99.999 %)	refined	2.32	4640	660
silicon carbide	sintered	3.15	4800	2827

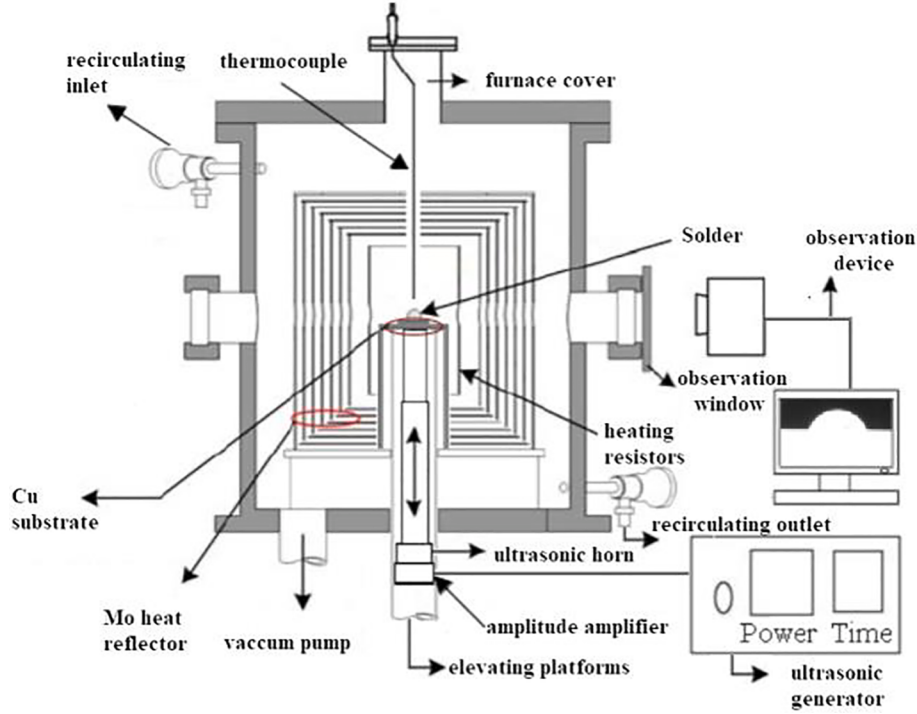


Fig. 1. Schematic of the ultrasonic device for wetting.

where energy terms include: the self-energy of an atom i named $E^{\text{self}}(r, q)$, the short-range interaction named $E^{\text{short}}(r, q)$, the Coulomb interactions named $E^{\text{Coul}}(r, q)$, the energies associated with atomic polarizability named $E^{\text{polar}}(r, q)$, the van der Waals interactions named $E^{\text{vdW}}(r)$, and the correction terms named $E^{\text{corr}}(r)$. Parameters of the Al–O potential have been trained for six typical aluminum oxide crystal structures: $\alpha\text{-Al}_2\text{O}_3$ (space group-R $\bar{3}c$), $\theta\text{-Al}_2\text{O}_3$ (space group $C2/m$), Al_2O_3 (Fe_2O_3 , space group $P4132$), AlO_2 (CaF_2 structure, space group $Fm\bar{3}m$), AlO_2 (TiO_2 , space group $P42/mnm$), and AlO (NaCl , space group $Fm\bar{3}m$) as given by Choudhary [36]. The kinetics of Al_2O_3 oxide film removal from Al droplets is determined using MD methods. The multi-resolution algorithms in use are of the Large-scale Atomic/Molecular Massively Parallel Simulator package [37].

2.2.2. A potential function for Al/SiC wetting process

In this part of the simulation, a vacuum environment is taken into consideration, after the Al_2O_3 oxides are removed, they spill out of the metal droplet and then draw into the vacuum. Therefore, before performing MD simulations, Oxygen atoms are excluded from the simulation models.

while the Al, Si, and C atoms still remain. This ternary spreading system is considered composite material, so a new hybrid potential is invoked. The Al–Al atomic pairs fit the embedded-atom-method (EAM) potential, and a tersoff interatomic force-field is adopted for SiC. The total potential energy terms (E_{total}) in EAM and the tersoff are separately expressed, using two different equations that are given below.

$$E_{\text{total-Tersoff}} = \frac{1}{2} \sum_i \sum_{j \neq i} \{ f_c(r_{ij}) [f_R(r_{ij}) + b_{ij} f_A(r_{ij})] \} \quad (2)$$

$$E_{\text{total-EAM}} = \sum_i F_i(\rho_i) + \frac{1}{2} \sum_{j \neq i} \Phi_{ij}(r_{ij}) \quad (3)$$

Inside Eq. (2), there are a cutoff function $f_c(r_{ij})$, the three-body interaction term $f_A(r_{ij})$, and the two-body interactive force $f_R(r_{ij})$. Meanwhile, $F_i(\rho_i)$ and $\Phi_{ij}(r_{ij})$ are the embedded energy and the core-core

repulsion energy for i th atom, or the i - j th atom pair. Symbols ρ and r_{ij} in Eq. (3) are their densities and the separation distances. The potential functions between Al–Si and Al–C are the simplest Lennard Jones potentials, whose parameters are: $\epsilon(\text{Al-Si}) = 0.482$ eV, $\epsilon(\text{Al-C}) = 0.469$ eV, $\sigma(\text{Al-Si}) = 0.391$ nm, and $\sigma(\text{Al-C}) = 0.342$ nm.

The melting and spreading simulations are carried out at a settled holding temperature. The constant value is 700 °C, which is controlled by the canonical ensemble at zero pressure. As is depicted in Fig. 2, a periodic displacement is applied for a wiggly layer of C atoms at the bottom, and the maximum amplitude of C atoms in motion during Al's melting process is fixed. All atoms at the bottom are kept moving in accordance with the sine-wave mode, and the paths of the substrate atoms are time-variant, as shown in Fig. 3. Each main area in MD analyses consists of 19,500 atoms that gradually transform from the solid into the liquid structure. Al and O atoms are distinguished by different colors in Fig. 2.

The simulation time step is set as 1 fs, and the whole simulation process lasts for 1,500,000 time steps. After finishing the MD simulations, several images of microstructures are obtained to determine the atomic motion. The radial distribution function relationships of Al and the surrounding atoms are described by the $g(r)$ - r curves. The curve shape varies with structural evolution as $g(r)$ can have diverse possibilities of finding atomic pairs within different given distances, R . Atoms are counted from 1 to N , $(1 \dots i \dots j \dots N)$ using the function given in Eq. (4),

$$g(R) = \frac{1}{N^2} \sum_i \sum_{j=i}^N \delta(R - |\vec{R}_{ij}|) \quad (4)$$

where $\langle \rangle$ denotes the average over the entire trajectory.

3. Results

3.1. Ultrasonically spreading processes

In Fig. 4, changing degrees of Al/SiC reactive wetting are shown with respect to time evolution. Fig. 4 (a) demonstrates the initial moment of

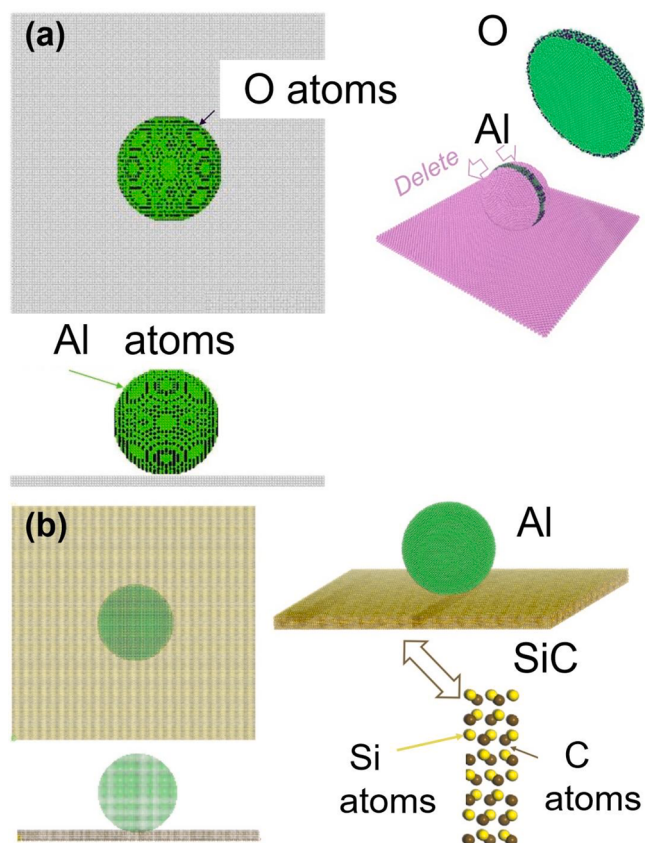


Fig. 2. Top, front, and perspective views of initial droplet/substrate atomic configurations in the simulations of sonoprocessing: (a) the oxide film structures built in numerical simulations (Al and O atoms are colored as green and black, respectively); (b) investigative procedure via MD with Al, Si, and C atoms colored in green, yellow, and brown, respectively.

solid–liquid contact, where the total ultrasonic excitation times of Fig. 4 (b), 4 (c), 4 (d), and 4 (e) are 3 s, 4 s, 5 s, and 10 s, respectively.

In most wetting situations, wetting is not accompanied with ultrasonic vibrations, Shen et al. have investigated the wettability of Al/SiC in wetting fields without ultrasonics [38]. Their results showed that Al_2O_3 becomes a big obstruction when the contact line is moving.

As revealed by wetting images, the flow of the sine-wave is allowed into the wetting area by the ultrasonic generator continually, and chromatic aberration is found in several images, which is colored in white at the static moments after 3–5 s. Ultrasonic vibrations aid the wetting system in eliminating the inhibition of oxide films soon. During the first 5 s, the substrate/liquid is still pinning the wetting field. The transient contact angle is 92.1° at 4 s and 87.6° at 5 s; however, the solid/liquid contact angle decreases at a faster rate in the next 5 s which corresponds to the ultrasonic spreading stage of 5–10 s.

The Al droplet is initially wrapped by the residual oxide film fragments. After exerting ultrasonic waves from the generator, the wetting process is then accompanied with the high power ultrasonics, so the sinusoidal ultrasonic wave pulls and pushes those fragments in a high frequency. In the meantime, it results in the quick decohesion of the oxide film. The process of chemical reaction can produce a series of substances, the thermodynamics of which are listed in Table 2, these chemical reactions between gaseous aluminum and the oxide film are greatly promoted and strengthened by the high intensity ultrasonic mechanical energy.

When ultrasonics are exerted into the wetting system, the acoustic cavitation bubbles have an impact on the interfacial area between oxide films and Al melts, the physical and chemical reactions are written as follows:

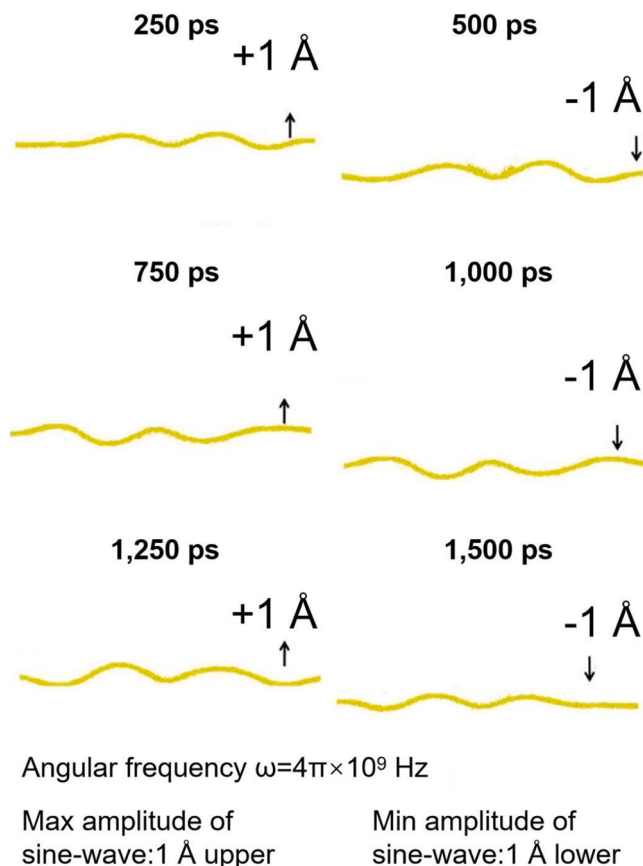


Fig. 3. Time-variant motion path of oscillating substrate atoms during the simulation of wetting under a sine-wave ultrasonic field.



According to Table 2, the possible reaction between Al_2O_3 and Al tends to be the reversible reactions. The fragment of oxide film acts as the raw material in chemical reactions. Laurent had ever point out the equilibrium oxygen pressure for Al/ Al_2O_3 , at 700°C , is alike to be 10^{-44} Pa . Here we could find that $P_{\text{O}_2}^{\text{eq}}$ is obviously lower than $P_{\text{Al}_2\text{O}}^{\text{eq}}$, which is $3.12 \times 10^{-13} \text{ Pa}$.

According to Le Chatelier's principle, once the pressure of gaseous reactant being elevated, the chemical system responds by pushing the positive direction of the reaction forward: $\text{Al} + \text{Al}_2\text{O}_3 \rightleftharpoons \text{Al}_2\text{O}(g)$ in Eq. (6), in order to lessen the change. So shell structure alumina structures are removed in a fast speed.

Numerical calculation demonstrates that ultrasonic bubbles cause transient local high temperature and high pressure, which separately reach 10,000 Kelvin and 10,000 times atmosphere separately [39]. Both high-level temperatures and pressures bring strong temperature shock and thermal stress shock to the oxide film. Since it produces a larger thermal stress, the thermal stress along with tensile stress breaks the shell structure Al_2O_3 quickly. The wettability of Al/SiC system could get further enhanced after the removal of oxide film by the effect of acoustic cavitation.

Except for the vibrational force which determines the degree of ultrasonic wetting, the extent of interfacial chemical reaction also takes effect. The effect of acoustic cavitation is crucial for the interfacial

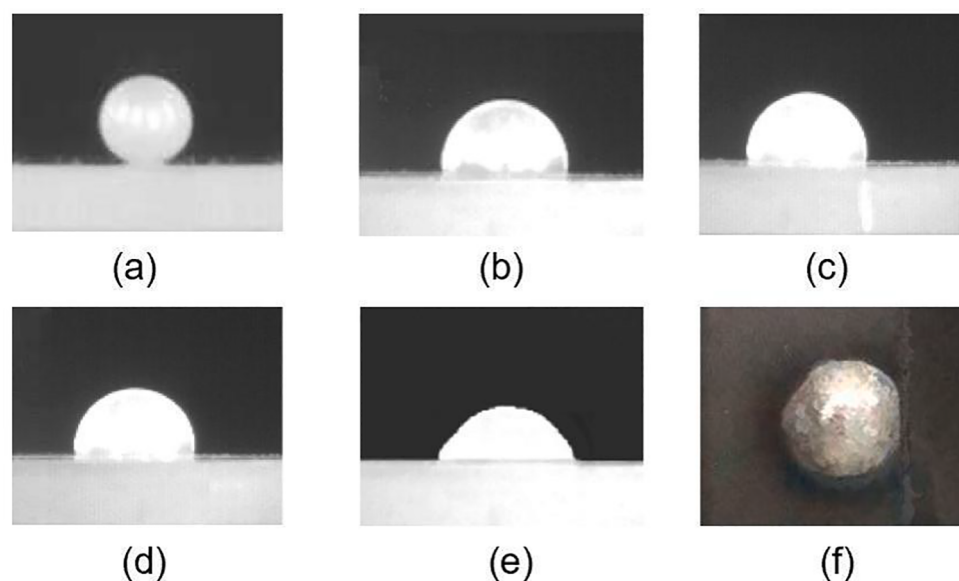


Fig. 4. Real images of molten Al wetting the SiC substrate in a 20 kHz ultrasonic field lasting for 10 s (a) at the initial stage, (b) 3 s later, (c) 4 s later, (d) 5 s later, (e) 10 s later, (f) the vertical view of Al/SiC sample.

Table 2

The chemical substances among possible reactions between oxide film and metal Al.

Substances	Al ₂ O ₃ (solid)	Al (gas)	Al ₂ O (gas)	AlO (gas)	AlO ₂ (gas)
Enthalpies of formation, kJ/mol	1669.68	329.7	-145.2	66.9	-86.2
Standard Gibbs free energy changes at 700 °C, kJ/mol	Raw material	Raw material	-1.56×10^{-6}	1.12×10^7	4.23×10^6
Saturated partial pressures of gases at 700 °C, Pa	Raw material	Raw material	3.12×10^{-13}	2.43×10^{-33}	2.13×10^{-32}
Activities	1.13 $l, m, n = 0, -4, 3$	1.051	1.10		
$a = a(\text{Al}_2\text{O}_3)^l a(\text{Al})^m a(\text{Al}_2\text{O})^n$		$l, m, n = -1, 0, 3$	$l, m, n = 1, 4, 0$		

reaction between Al and SiC. After propagating 20 kHz ultrasonic waves into the melt, micro-jets are generated by the ultrasonics, and the cavitation bubbles collapse, hit, and broke in molten Al [40]. Since there are instantaneous high temperature and high pressure inside the melt, very strong temperature shock and thermal stress shock are given to the oxide film. Also, the high temperature together with the high pressure

significantly promote the interfacial chemical reaction, in turn, resulting in producing a larger precursor film's area. During the enlarging process of the precursor film, the dissolution and diffusion continue speedily between molten Al and SiC substrate because of the acoustic cavitation effect. The above-mentioned suggest that the wettability can be enhanced by the effect of acoustic cavitation.

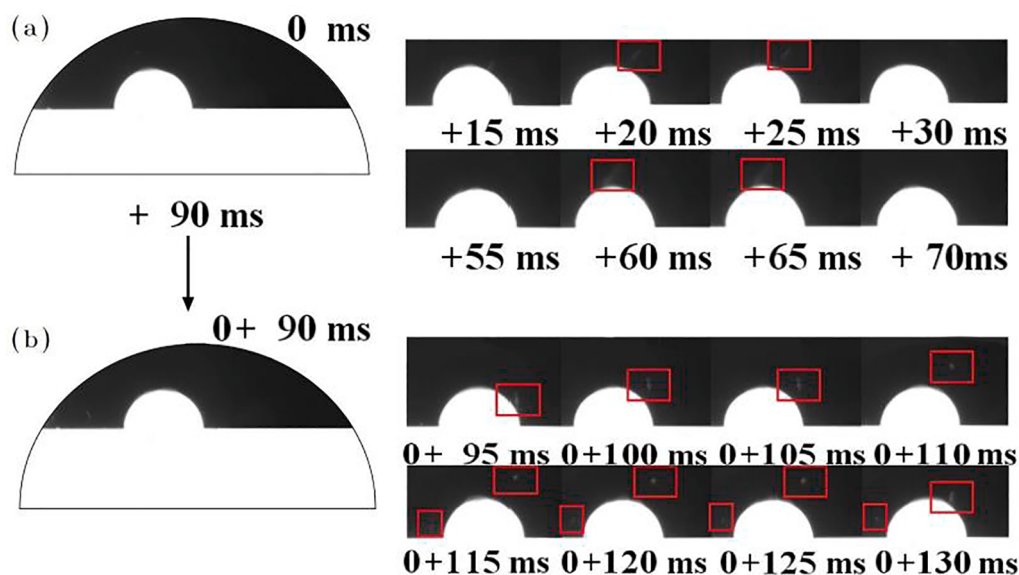


Fig. 5. Images of dynamic wetting behavior in Al/SiC, with a short period of ultrasonic activation: (a) within time periods from 15 to 30 ms and 55 – 70 ms, (b) within the period from 90 to 130 ms.

3.2. Removal of molten aluminum oxide by ultrasonics

3.2.1. Experimental observations

Without removal of Al_2O_3 films, the molten Al droplet is wrapped around. The liquid metal beneath the substrate is pinned and prevented from unfolding quickly. During the photography of the ultrasonic spreading process, no characteristics of enlarging contact area are seen for the system of Al/SiC (with Al_2O_3 outer surface). For the initial period of 130 ms, the dynamic process is shown in Fig. 5.

Roughly 5 s later, the droplet becomes free of contact line pinning. We can judge from the video that most tiny perturbations are replaced by shape oscillations in that respective moment. These dynamic characteristics can bring the vital signal of molten Al's free motion during spreading. Sonoprocessing by ultrasonics has led molten Al droplets to have less time struggling with the oxide film barrier.

As shown in Fig. 5 (b), the total time period of using ultrasonic waves to eliminate Al oxidation is shorter than 5 s. In contrast with the Al/SiC system in commonly non-ultrasonic wetting fields, the time frame of oxide removal has significantly shortened. When molten liquid no longer struggles with Al_2O_3 , the perturbation of molten Al's droplet shape becomes more evident.

3.2.2. MD investigations of deoxidation processing

The side view of atomic microstructure cross-sections of Al on SiC substrates is shown in Fig. 6. Through Fig. 6 (a–f), the actions of oxygen atom removal out of the inner region of the metal are demonstrated in sequential order. In Fig. 6 (a, b), the spreading process with ultrasonics follows the first full circle of the megacycle hertz wave. During this circle

of megacycle hertz waves, light green metal atoms are still arranged at the sites of FCC superlattices. We pay close attention to the locations of Al-oxides surrounding the metal core. The microstructure of the atomic system show no configurations of melting. Before 500 picoseconds, the fluidic motion is fully restrained by the outer shell wrapping Al. With the adaptive common neighbor analysis, various types of transitional structures are quantitatively estimated. The column charts are depicted below, that corresponds to the atomic structure change at each moment.

For the next ultrasonic wetting period, the moments are demonstrated by wetting images in Fig. 6 (c, d). In this process, the molten metal atoms do not remain enclosed. As the patterns demonstrate the oxide structures become scattered along the outlines where the oxide film was supposed to be ruptured. In Fig. 6 (e), it is found that oxygen atoms are scattered outside the metal Al area, and the distribution of the Al–O structure is not uniform. After 1250 ps, the metal Al inside the shell fully transitions into the liquid phase, as shown in Fig. 6 (f). The droplet contour line also changes during wetting moments shown in Fig. 6 (e, f). The Al droplet appears to be in an inverted pyriform shape. The whole process in Fig. 6 shows melted aluminum pouring out of the shell when treated with ultrasonics during the heating process.

3.2.3. MD investigations of sonoprocessing of Al/SiC wetting

During dynamic wetting without ultrasonics as the contrast process, there are six key nodes shown by wetting triple-line morphology drawn on the light blue background (Fig. 7). When no ultrasonic vibration is induced in the wetting system during MD simulations, the contact angle varies slowly. The interval between every-two moments being captured is 250 ps. At 0 s, the contact angle by MD is obtuse, which is illustrated at the beginning of the second row. Over time, the aluminum atoms with triple-line motion inside the MD model move outwards, so the droplet region is in contact with the SiC outer surface in a larger area. The horizontal instantaneous velocities at different time points have been calculated according to the scale bar and the outline information. The contact angle data are 84.4°, 80.2°, 77.6°, 74.7°, 72.5°, and 71.2° at 250 ps, 500 ps, 750 ps, 1000 ps, 1250 ps, and 1500 ps, respectively.

By contrast, the contact angle decreases at a relatively fast rate when substrate atoms are moving with a wiggle oscillation. During the simulation of sonoprocessing at 500 ps, 750 ps, 1000 ps, and 1250 ps, the values of the horizontal velocity of Al atoms near the triple line are 0.041 Å/ps, 0.044 Å/ps, 0.041 Å/ps, and 0.044 Å/ps, respectively. Without the oscillating substrate atoms, the Al atoms become slow-moving, whose velocities are 0.019 Å/ps, 0.022 Å/ps, 0.031 Å/ps, and 0.038 Å/ps. The spreading process is significantly enhanced, terminated by the input of ultrasonic oscillation. Meanwhile, the minimum contact angle is finally down to 46.7°, which is much lower than the results of the contrast process. The contact angle is reduced by 34 percent when compared with the MD simulation of non-ultrasonic treatment. The precursor film characteristic appears after 250 ps of sonoprocessing treatment, whose structure is made up of the blue Al atoms. Once the vibration process comes into the positive semicircle, a stronger growing tendency of atomic vertical motion promotes the atomic mobility of Al. Since the Al atoms are involved in interfacial diffusion, the dissolution reaches a higher level, and the pink atoms inside the substrate's diffusion layer increase in quantity. The rule indicated by the observed liquid/substrate configurations inside Fig. 7 is that as the ultrasonic wave peaks, the rate of convective diffusion processes of Al, Si, and C atoms also increases, which leads to more Al atoms being absorbed inside the diffusion layer. Thus, the precursor film structure also becomes significant. The level of wettability gets promoted, and lower transient contact angles are seen.

4. Discussions

4.1. Liquids structural transition

In order to deeply understand the interaction between atomic

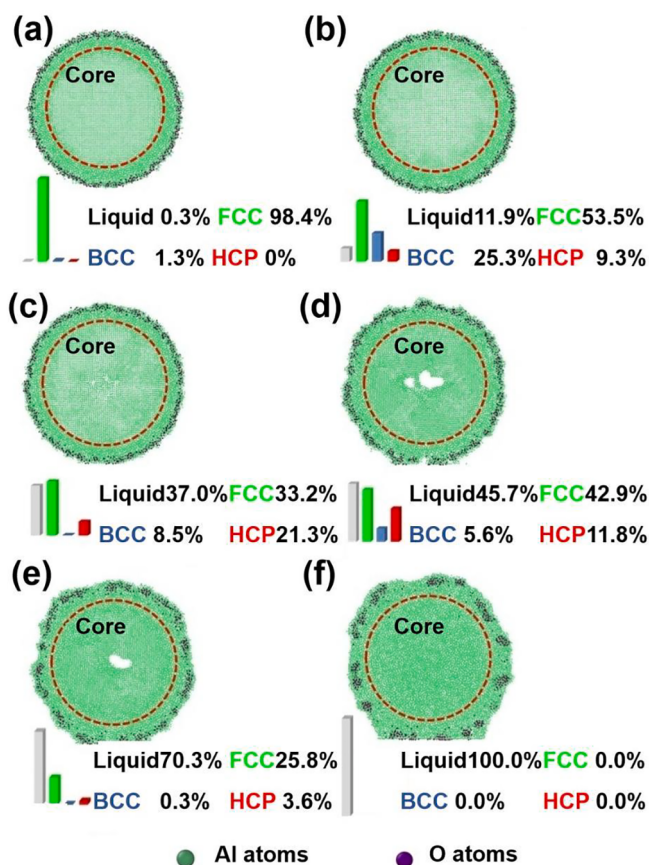


Fig. 6. MD images of Al droplet during the different stages of ultrasonic spreading in sectional views attached with the ratio of different atomic structures, including liquid, face-centered cubic (FCC), body-centered cubic, and hexagonal close-packed, after the ultrasonic spreading periods of (a) 250 ps; (b) 500 ps; (c) 750 ps; (d) 1000 ps; (e) 1250 ps; and (f) 1500 ps.

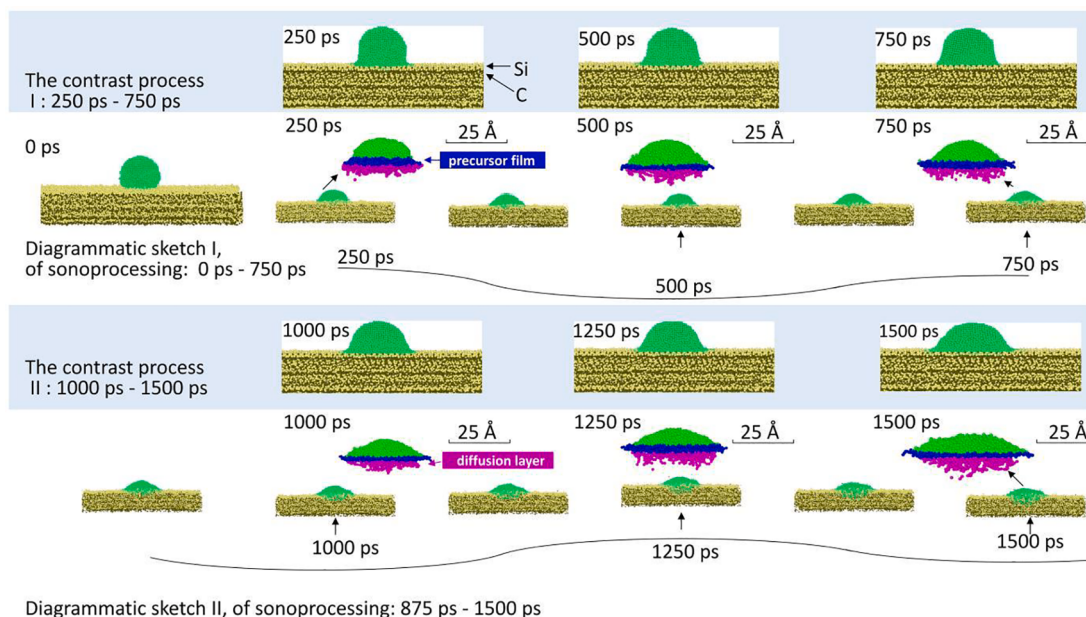


Fig. 7. MD images of Al wetting on SiC in sonoprocessing. The whole spreading period lasts for another 1500 ps, composed of three ultrasonic cycles. The images with the light blue background reveal the contrast process.

structure and the periodical ultrasonic stage, Fig. 8 demonstrates the variation of RDF at different ultrasonic spreading moments. As demonstrated by the results, the metal structure in the MD model first remains solid, then shows an inversion tendency, and finally accomplishes phase transition. In the middle stage of the ultrasonic period, i.e., from 750 to 1000 ps, the secondary and tertiary peaks of RDF vanish. In the light of crystal structure percentages calculated in Fig. 6, the liquid atoms have replaced the atoms at FCC structural regions. At 1000 ps, several microvoids appear from the center of the droplet. At that moment, the molten droplet volume increases rapidly, and the microstructure void area is produced in order to balance the variation of Al density. Afterwards, the RDF curve attains a uniform shape. As long as the Al structure remains fully liquid, the oxide film does not affect the fluidity of liquid Al. Therefore, the additive ultrasonic force takes full effect.

4.2. Analysis of theoretical models

4.2.1. Ultrasonic radiation forces

As is shown in Fig. 4, the contact angle value is first 0.833π radian, then 0.487π radian (after oxide films are totally removed), and finally 0.395π radian. Thus, the wetting degree between Al/SiC has already been enhanced. During the whole 10 s of the ultrasonic treating process, there must be an additional impetus for the wettability enhancement. As can be deduced that ultrasonic radiation forces inside the molten Al region take effects, so we project the following statements, in order to quantitatively prove the existence of ultrasonic radiation force.

Considering the linear and non-linear physical phenomena of ultrasonic wave propagation in a liquid, the acceleration is written as follows:

$$\frac{du}{dt} = \frac{\partial u}{\partial t} + (u \cdot \nabla)u \quad (9)$$

The second term on the right symbolizes the convective acceleration; it cannot be ignored since the wave intensity I in this three-

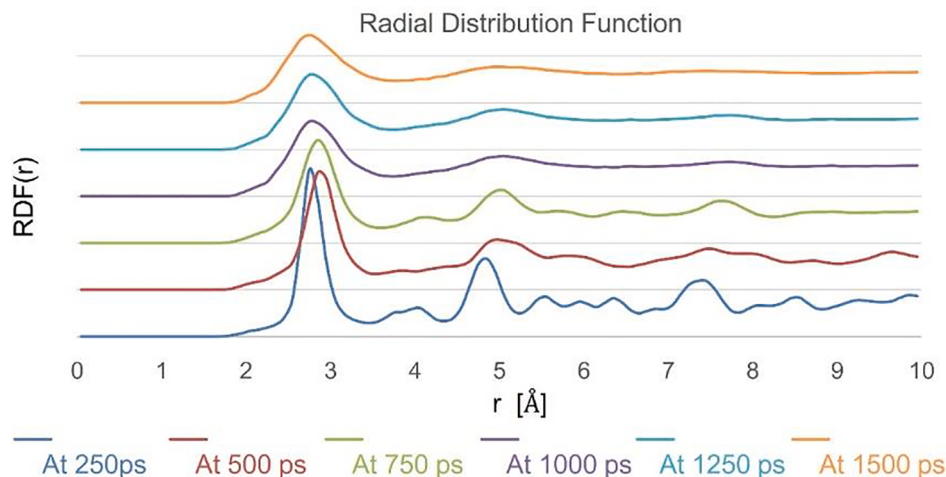


Fig. 8. MD simulation results of Al droplet microstructure indicated by different RDFs during the ultrasonic spreading process. The curves are depicted separately after 250 ps; 500 ps; 750 ps; 1000 ps; 1250 ps; and 1500 ps.

dimensional ultrasonic field is overlage. The acoustic pressure p , as well as the impedance term $\rho^{Al} \cdot c^{Al}$, contributes to the intensity of ultrasonic waves in Eq. (10).

$$I = \frac{dW}{Adt} = \frac{p^2}{\rho^{Al} \cdot c^{Al}} \quad (10)$$

The biggest contact area of liquid/substrate is $3.309 \times 10^{-5} \text{ m}^2$ and the estimated value of I is high. The added energy for wetting molten Al is an output of the acoustic source. The wave velocity deeply relates to the droplet of motion: $\partial u / \partial t = (\omega \cdot a) \partial \cos[2\pi z(t)/\lambda - \omega t] / \partial t$, where ω , a , λ , and t are ultrasonic attributes, namely the angular frequency, the amplitude, the wavelength, and the excitation time. z , on the other hand, demonstrates the tip to height, as marked in Fig. 9.

For any type of body force, the height of a droplet's center is z' , $z'(t) = z(t) - R_t \cos \theta_t$. Here, R_t is the corresponding radius of the sphere crown. Therefore, we refer to the equation of the center of gravity: $z = \iiint z \rho dx dy dz / \iiint \rho dx dy dz$. The relational expression of $z'(t)$ is investigated in the next part. Droplet volume V is constant in principle, while the time-variant contact area on the solid surface is symbolized by A . Geometrical relationships between V , A , and θ_t , R_t , can be expressed as Eqs. (11) and (12).

$$V = \left(\sqrt{\frac{A^3}{\pi}} / 3 \right) (2 + \cos \theta_t) (1 - \cos \theta_t)^2 \sin^{-3} \theta_t \quad (11)$$

$$R_t = \sqrt[3]{3V/\pi} (1 - \cos \theta_t)^{-2/3} (2 + \cos \theta_t)^{-1/3} \quad (12)$$

The ultrasonic output stress P_{output} functions as the ensemble of (W , t , c , ρ , V , and θ_t),

$$P_{output} = \sqrt{W \rho^{Al} c^{Al} / A t} \quad (13)$$

For which, the differential equation to variant t is as Eq. (14):

$$dP_{output} / dt = - \left(\sqrt{W \rho^{Al} c^{Al} / 2} \right) t^{-3/2} \quad (14)$$

4.2.2. Thermodynamics of spreading with ultrasonics

For a spreading Al droplet, the dynamic degree of variations in intensity can be ignored as time goes on, considering that the dynamic spreading process is no longer than 10 s. During this period, A and R_t (R_t equals $r_t / \sin \theta_t$) increase while $z(t)$ decreases. The spherical crown's shape variation is shown in Fig. 10.

Then, the center of gravity $z(t)$ is calculated, and the variants of R_t , $z(t)$, and $z'(t)$ are successfully rewritten by θ_t and t .

$$z(t) = \frac{\Delta z \rho dx dy dz}{\Delta \rho dx dy dz} = \frac{3}{4} \sqrt[3]{3V/\pi} \sin^4 \theta_t (1 + \cos^2 \theta_t) (1 - \cos \theta_t)^{-8/3} (2 + \cos \theta_t)^{-4/3} \quad (15)$$

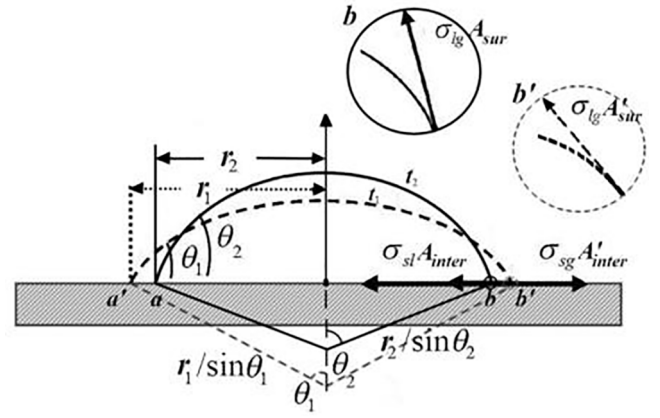


Fig. 10. Schematics of the dynamic hemisphere model. The moving contact line varies from ab to $a'b'$ while the contact angle and A vary from θ_2 to θ_1 and from $r_2/\sin \theta_2$ to $r_1/\sin \theta_1$, respectively.

$$z'(t) = \sqrt[3]{3V/\pi} [3 \sin^2 \theta_t (1 - \cos^4 \theta_t) (1 + \cos \theta_t)^2 / (8 + 4 \cos \theta_t) - \cos \theta_t] (1 - \cos \theta_t)^{-2/3} (2 + \cos \theta_t)^{-1/3} \quad (16)$$

The classical Young's equation is shown in Eq. (17),

$$\sigma_{lv} \cos \theta_t = \sigma_{sv} - \sigma_{sl} \quad (17)$$

When it comes to surface thermodynamics, it arrives that $dG = \sigma_{lv} dA_{sur} - (\sigma_{lv} \cos \theta_t) dA_{inter}$, $dA_{sur} = 4\pi R_t (1 - \cos \theta_t) d\theta_t$, $dA_{inter} = 2\pi R_t \sin^2 \theta_t d\theta_t$, $G = \int [2/(1 + \cos \theta_t) - \cos \theta_t] \sigma_{lv} dA$. When in kinetics, the A_{inter} in Fig. 10 is also marked as A , stating the increment of body forces are always conserved when in equilibrium, $dA/dt = 0$.

$$\int_A P_{ultrasonic} [\delta z'(t)] dA = \int_A [2/(1 + \cos \theta_t) - \cos \theta_t] \sigma_{lv} dA \quad (18)$$

The differential of Eqs. (19) and (20) are separately:

$$\frac{dz'(t)}{dt} \approx - \frac{2\sqrt[3]{3V/\pi} \sin \theta (\cos^2 \theta + 1)}{\sqrt[3]{\pi} (1 - \cos \theta)^{5/3} (2 + \cos \theta)^{4/3}} \frac{d\theta}{dt} \quad (19)$$

$$- 2\sqrt[3]{3m^{Al}/\rho^{Al}} \frac{\cos^2 \theta + 1}{\sqrt[3]{\pi} (1 - \cos \theta)^{5/3} (2 + \cos \theta)^{4/3}} dP_{ultrasonic} / dt = \sigma_{lv} \left[1 - \frac{2}{(1 + \cos \theta)^2} \right] \frac{dA}{dt} = 0 \quad (20)$$

The dimensionless symbols X_θ and Y_θ are defined to make easier quantitative estimations of the above-mentioned $P_{ultrasonic}$. X_θ equals $(\cos^2 \theta + 2 \cos \theta - 1) \cdot (1 - \cos \theta)^{5/3} \cdot (2 + \cos \theta)^{4/3}$, while Y_θ

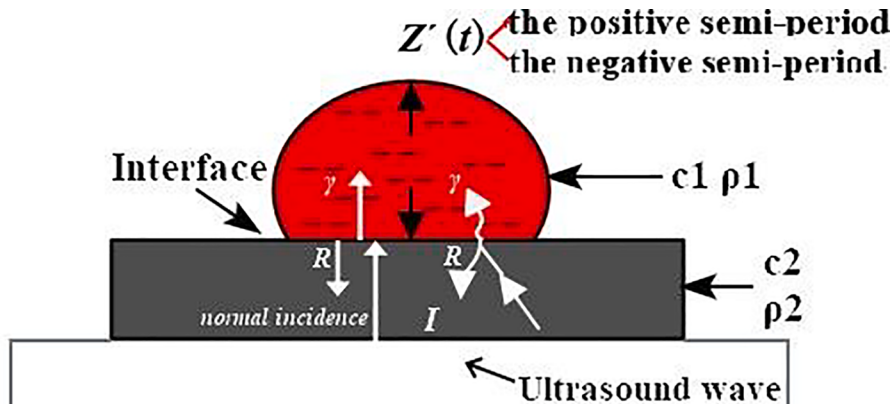


Fig. 9. Schematic of ultrasonic energy propagation in Al/SiC ultrasonic spreading system.

equals $(\cos^2\theta + 1) \cdot (1 + \cos\theta)^2$.

4.3. Thermodynamics of ultrasonic spreading

4.3.1. Contact angle of Al on SiC during sonoprocessing

Within the first 5 s of ultrasonic spreading, the oxidation film around the molten Al droplet is already removed. The images of Al/SiC equilibrium wetting states are continuously captured from 5 – 10 s. There are 26 in situ wetting moments that are shown in Fig. 11. Here, the black-and-white photographs are from a non-stop video of the Al/SiC spreading process in the 20 kHz ultrasonic field. This is more convenient for investigating the dynamic variation of solid/liquid macroscopical contour, which is further interpreted into the relationship between the contact angle θ and the ultrasonic excitation time t .

The contact angle as a criterion of wettability is also estimated in kinetics. Images in Fig. 11 show that the contact angle decreases at a significantly faster rate. Thus, using ultrasonic waves for the wetting process is crucial to enhance Al/SiC wettability.

The function of $f(\theta) - t$ is also related to the thermodynamic basics of Al/SiC wetting. Dezellus [41] has first provided the kinetic model of spreading, where k equals a constant value and the three subscripts of θ from left to right mean that the respective contact angles are instantaneous, equilibrium, and initial.

$$\cos\theta_t = \cos\theta_E - (\cos\theta_E - \cos\theta_i)\exp(-kt) \quad (21)$$

Based on the chemical reaction-limited spreading kinetics, Zhang et al. [42] have also used the exponential relationship to predict the solid–liquid interfacial energy, see Eq. (22), where c is the atomic concentration in the reaction process. In the wetting laws of Dupre, the work of adhesion is expressed as $W = \sigma_{SL}(1 + \cos\theta_t)$. During the spreading process demonstrated in Fig. 11, the solid–liquid contact line keeps extending outwards, and the time-variant thermodynamic phenomena indicate that the Al/SiC system is accumulating the adhesion increasingly.

$$\sigma_{SL} = (\sigma_{SL}, I - \sigma_{SL}, E)\exp(-kct) + \sigma_{SL}, E \quad (22)$$

Where c is the atomic concentration in the reaction process, besides, in wetting laws of Dupre, the work of adhesion expresses as $W = \sigma_{SL}(1 + \cos\theta_t)$. During spreading process demonstrated by Fig. 11, the solid–liquid contact line keeps extending outwards regularly, and those time-variant thermodynamic phenomena indicate that Al/SiC system is accumulating the adhesion work increasingly.

4.3.2. Surface thermodynamics

The first law of thermodynamics is:

$$\Delta U = Q + W \quad (23)$$

when molten metal ultrasonically spreads, the ultrasonic waves

allow the energy W into the solid/liquid system. As it has been deduced before, P_{Input} varies with the value of solid–liquid interface area A ; thus, $dW = P_{\text{Input}}(t)dA$. Only the partial work symbolized by variable $W_{\text{Ultrasonics}}$ can promote the wettability of Al/SiC from 5 – 10 s. Actually, $W_{\text{Ultrasonics}}$ also depends on effective power $P_{\text{Ultrasonics}}$, see Eq. (24). Undoubtedly, estimating the percentages of efficiency in different ultrasonic fields might be of an appreciable engineering value. With the mathematical relationships from Eqs. (11) to (22), we have managed to quantify the exact efficiency values of ultrasonic wetting with η_{Eff} . For the efficiency of Al/SiC wetting, from 5 to 10 s under 20 kHz ultrasonic field, the numerical solutions are as follows.

$$P_{\text{Ultrasonics}} = \int \frac{\sqrt[3]{\pi/3}}{2} \sqrt[3]{\rho^{\text{Al}}/m^{\text{Al}}} \sigma_{\text{lv}} X_{\theta} dt, P_{\text{Input}} = \int \sqrt{W \rho^{\text{Al}} c^{\text{Al}} / A} dt, \eta_{\text{Eff}} = P_{\text{Ultrasonics}} / P_{\text{Input}} \quad (24)$$

Table 3 lists the calculated values of time-variant thermodynamic parameters such as P_{Input} and $P_{\text{Ultrasonics}}$. $P_{\text{Ultrasonics}}$ jumps from nearly $300 \text{ N}\cdot\text{m}^{-2}$ to $1.656 \text{ N}\cdot\text{m}^{-2}$ within another 5 s. At 10 s, the contact angle is already the lowest, 0.395π radian. From Table 3, the cosine of contact angle of ultrasonic spreading can be described by a parabola for its increasing trend. After 10 s, the wetting field already has sufficient vibrational energy. Then, the spreading of liquid droplets meets obstructions from the equilibrium balances of the interfacial energy. P_{Input} is also a comprehensive variable but not as sensitive as $P_{\text{Ultrasonics}}$.

As we have already evaluated the varying dynamic trends of X_{θ} and Y_{θ} , it is proved that the thermodynamic variables P_{Input} and $P_{\text{Ultrasonics}}$ are determined by the contact angle θ , as well as ultrasonic excitation time t . In Fig. 12 (a) and (b), P_{Input} and $P_{\text{Ultrasonics}}$ decrease exponentially from 5 to 10 s. Fig. 12 (c) indicates that the dynamic ratios of $P_{\text{Ultrasonics}}/P_{\text{Input}}$ are in a chronological logarithmic descending trend. The quotient demonstrates how much the effective energy for wetting has evolved in the equilibrium spreading process, corresponding to the variable η_{Eff} in Table 3. As already revealed by $P_{\text{Ultrasonics}}$, P_{Input} , and η_{Eff} in Eqs. (12–18) are all time-dependent. The curve demonstrates the accumulation value of effective ultrasonic work. Its divergence to the linear line shows a parabolic trend, as shown in Fig. 12 (d), it reaches the farthest deviation point at about 7 s. The integrated energy of ultrasonics reaches its saturation value when the spreading process ends. During the experimental tests, the equilibrium contact angle in the 10 s of the ultrasonic spreading process is 65.9° .

5. Conclusion

The method of improving the wettability of Al droplets on SiC ceramics has been investigated in this paper. A continuous ultrasonic spreading process accomplishes this within 10 s. In the dynamic ultrasonic wetting process, precursor films can emerge quickly with the assistance of ultrasonics. It is proved by dynamic experiments and

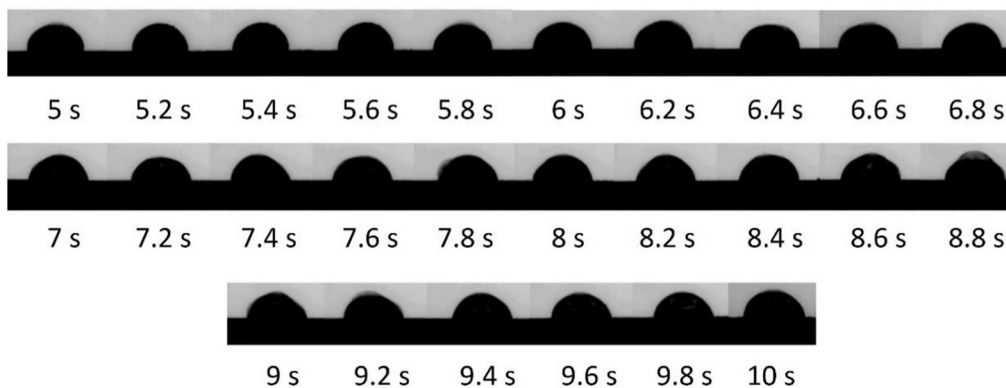


Fig. 11. Ultrasonic wetting images of Al/SiC at moments from 5 s to 10 s.

Table 3

Calculated values of specimen materials.

Ultrasonic excitation time	5 s	6 s	7 s	8 s	9 s	10 s
$\cos\theta$	0.041	0.173	0.272	0.335	0.381	0.407
X_{θ} , Dimensionless	733.150	324.167	139.444	62.024	22.155	4.055
Y_{θ} , Dimensionless	0.045	0.034	0.027	0.022	0.019	0.016
$P_{\text{Ultrasonics}}$, N/m ²	299.382	132.374	56.942	25.328	9.047	1.656
P_{Input} , N/m ²	6561.951	4991.845	3961.329	3242.299	2717.216	2320
η_{eff} , dimensionless	—	0.035	0.029	0.025	0.021	0.019

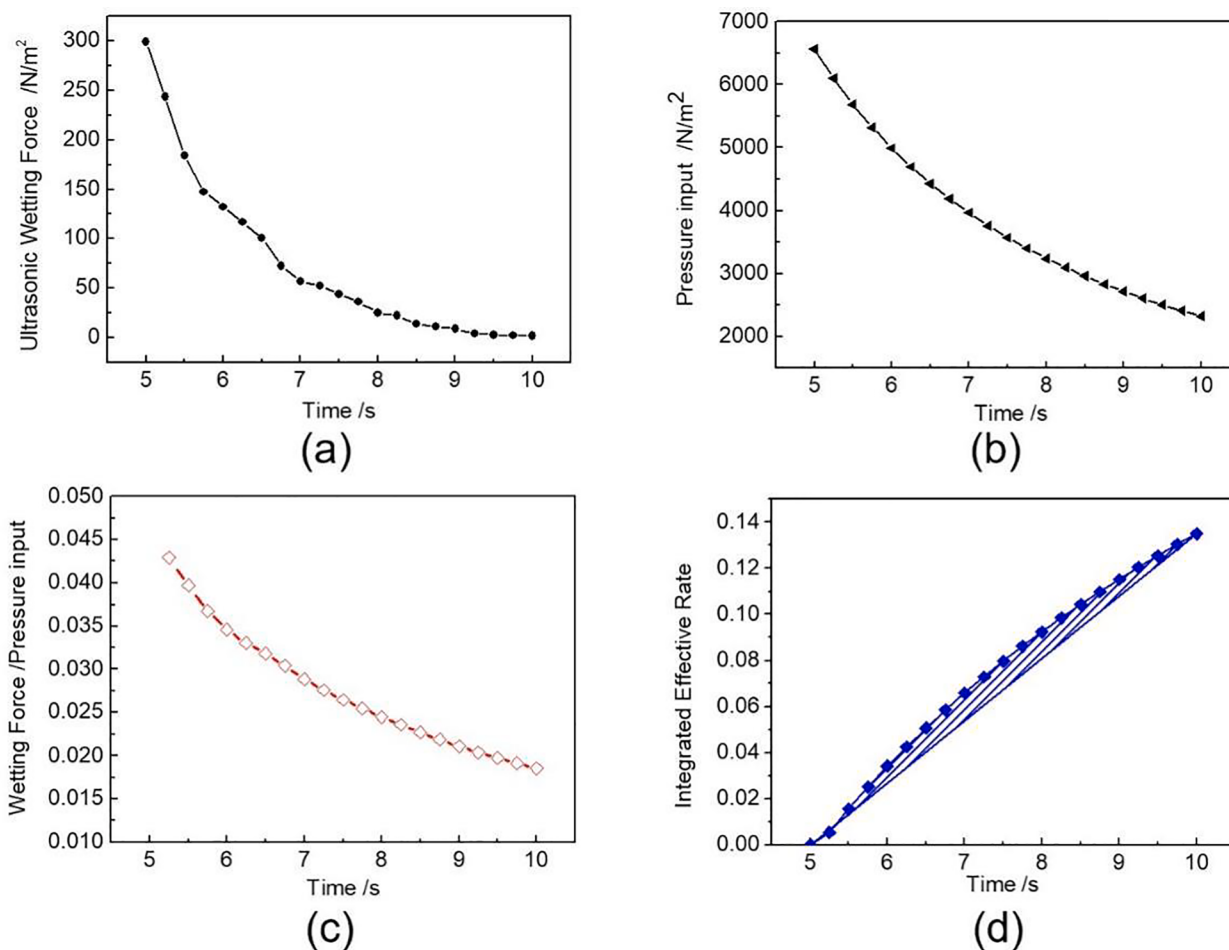


Fig. 12. Charts of thermodynamic powers: (a) $P_{\text{Ultrasonics}}$, and (b) P_{Input} versus ultrasonic excitation time, along with: (c) the time-variant efficiency, η_{eff} , and (d) integrated rates of the ultrasonic force to the wetting force.

numerical calculations that chemical and physical changes on either the surface or the interface determine the fast-evolving solid/liquid wetting behavior, as well as the production of precursor film on the SiC substrate. The appearance of precursor film can enhance the spreading rate of Al droplets on the SiC ceramic surface. The dynamic wetting behavior of Al/SiC has been revealed by thermodynamic calculations and MD simulations. Through the analysis of high-speed photography, it was found that the Al_2O_3 film inhibits a higher wetting degree of the Al metal droplet. The removal of oxide film begins at 3 s and ends at 5 s in the entire ultrasonic period. Unaffected by oxidic structures, the contact angle rapidly decreases to 87.6° , turning from an obtuse contact angle into an acute angle, which represents that the Al droplet spreads spontaneously. The precursor film starts to emerge in the spreading process. MD simulation indicates that the O atoms located in the shell unwrap the Al atoms in the core. When periodic vibrational force becomes active on the shell structure, the oxide film first breaks from where the O atoms are relatively few. The Al microstructure gets better flowability with the

existence of sine-wave motions of substrate atoms, and the wettability of Al/SiC is enhanced

The kinetic rules below are indicated by MD simulations. When a high-frequency motion is introduced by ultrasonic oscillation, the speed of horizontally moving Al atoms near the solid/liquid interface doubles. At 500 ps, the instantaneous outward speed reaches $0.041 \text{ \AA/ps}$, which is 2.17 times as large as the speed of atoms in non-ultrasonic wetting. Ultrasonic spreading gives impetus to the atomic motion both toward the exterior region on the substrate and evolving into the solid/liquid dissolution inside the diffusion layer. At 1500 ps, the contact angle value is 71.2° with no ultrasonics during the wetting simulation, while it falls to 46.7° (a decrease of 34 %) as the ultrasonics are being introduced in the simulation process. In short, adding ultrasonic waves determines the production of precursor film structure and also immensely activates liquid spreading while enhancing the liquid wettability. The kinetic law of spreading, forming the precursor film, and reducing contact angles are well fitted with the experiment results.

This paper analyzes the thermodynamics of ultrasonic spreading. The decrease of dynamic contact angle θ becomes the key factor to variations in both thermal and ultrasonic energies in wetting. For the time period from 5 to 10 s, the numerical expressions for increments of the input and the effective energy powers, namely P_{Input} and $P_{Ultrasonics}$, have been separately evaluated in this paper. According to the calculations, P_{Input} decreases from $6561.951 \text{ N}\cdot\text{m}^{-2}$ to $2320 \text{ N}\cdot\text{m}^{-2}$ (a reduction of 65 %). The effective surface energy power, $P_{Ultrasonics}$, decreases from $299.382 \text{ N}\cdot\text{m}^{-2}$ to $1.656 \text{ N}\cdot\text{m}^{-2}$ (a reduction of 99.5 %). According to the wetting kinetics approximation, the efficiency η_{Eff} is in a logarithmic descending trend as the wetting contact angle also decreases gradually from 87.6° to 65.9° .

CRediT authorship contribution statement

Wendi Li: Methodology. **Yuxin Liang:** Formal analysis, Data curation, Writing - review & editing. **Bangsheng Li:** Funding acquisition, Resources, Software. **Jicai Feng:** Conceptualization, Supervision.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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