Original Article

Wetting of liquid aluminum alloys on pure titanium at 873–973 K

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The wetting behaviour of Al alloys (i.e., 6061 Al and 4043 Al alloys) on the surface of pure titanium was studied by sessile drop method at temperatures of 873–973 K under high vacuum with \(10^{-4}\) Pa. All wetting processes were divided into two stages: in the nonlinear stage, the formation of a compact intermetallic layer (\(\text{Ti}_7\text{Al}_3\text{Si}_{12}\)) blocked the mass transfer at solid/liquid (S/L) interface and deteriorated wetting, while the formation of loose phase (Al\(_x\)Ti) at triple line region can removed the oxide film and improved wettability. The wetting activation energy (56 kJ/mol for a 6061Al/Ti system, and 71 kJ/mol for a 4043Al/Ti system) corresponded to the sum of the difference in surface energy per molar area between \(\alpha\)-Ti and TiO\(_2\) (27.2 kJ/mol), and the Gibbs energy of formation for Al\(_x\)Ti (30 kJ/mol) or the decomposition energy for Ti\(_7\)Al\(_3\)Si\(_{12}\) (48 kJ/mol). In the near-linear stage, the droplet spread on the surface of intermetallic compound (IMC). The spreading dynamics for 6061 alloy satisfied the Jiang’s empirical equation, and for 4043 alloy satisfied the viscous dissipation-limited model. The interfacial reaction had little effect on a droplet spreading, while the viscous friction drove the spread of 4043 alloy on precursor film in the near-linear stage.

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1. Introduction

Aluminum-titanium components have been widely used in petrochemical components, as biocompatible materials, and in aircraft engines due to their light-weight nature, oxidation resistance, and corrosion resistance [1,2]. Developing such aluminum-titanium components with excellent tensile strength allows their substitution for aluminum, as used in high-speed wind tunnels and propulsion exhaust systems to improve corrosion and oxidation resistance at high temperatures [3]; however, the joining process requires lower heat input to avoid residual stress because of their weakly metal-
lurgical compatibility and the mismatch in chemical affinity [4]. Besides, the brittle Ti-Al IMCs are considered to be a source of cracking which causes joint fracture [5]. To decrease the heat input and avoid the formation of thick Ti-Al IMCs at Ti/Al joints, many researchers pointed out that the addition of trace elements can modify the phase compositions, especially for the Mg and Si [6,7]. Chen et al. [7] reported on the microstructure of Ti to Al joining by laser, and found that the heat input was decreased, the Ti-Al IMCs layer thinned and the strength of the joint increased; Si element segregation at solid-liquid interface affected the microstructure and tensile strength. Gao et al. [8] described how the hybrid heat sources was key to determining the joint strength rather than the type of groove in a Ti-Al joining process using laser-arc hybrid welding. Li et al. [1] revealed the addition of elemental Mg into Al filler wire can significantly improve the joint strength and further inhibit the precipitation of Al-Ti IMCs. Cheng et al. [9] studied the vacuum brazing of Ti and Al, and indicated that with the temperature increasing, the wettability of solder was gradually improved. The Mg and Si elements can effectively reduce the thickness of Ti-Al IMCs, and improve joint strength. Besides, trace amounts of Mg can reduce the Ti2O and Al2O3 oxide film [10], and the addition of Si can contribute to a good flowability because of the surfactivity of Si in molten Al-Si alloy [11]. Therefore, suppressing the formation of Ti-Al IMCs and optimising the interfacial structure are of great concern to improving joint strength. However, heat input was not suited to being decreased to a certain value given the poor wetting behaviour and joint appearance of Ti/Al joints, limiting the further improvement of their joint properties. The intrinsic mechanism of interfacial microstructure regulation and joint formation is closely related to wetting and interfacial reaction behaviour of the Ti-Al metallurgical system with small amount of Mg and Si thus justifying this work.

Recently, some researchers have conducted preliminary studies of the wetting behaviour of molten metal on the surface of Ti plate. Liu et al. [12] reported on the wettability of liquid Ti32.3Zr0.7Ni3.3Cu6Be27.7 on the surface of TC4, and indicated that as the temperature increases, the improvement of wettability was related to the reduced viscosity and surface tension of molten metal, besides the interfacial reaction could promote the wettability. Tashi et al. [13] found a temperature dependence in the wetting of AgCuZn to TC4. Lin et al. [14] suggested that the spreading dynamics of an Al drop on steel plates was independent of the interfacial reaction and determined by the characteristics of droplet itself. The wettability and spreadability were gradually improved with the increase of heat input and the capillary force was the factor limiting droplet spreading. Zhou et al. [15] investigated the wetting of 4043 Al alloy on galvanized steel, and found that the spreading of liquid Al was limited by the viscosity. Li et al. [16] claimed that the oxide film blocked the moving of triple line, while the interfacial reaction could remove the oxide film, making the surface of substrate to be more metallic, and enhance the affinity between droplet and metallic substrate. Eustathopoulos et al. [17] introduced a reaction product control model for a metal/ceramic system, and claimed that the production of intermetallic compounds can effectively promote wetting. Satyanarayana et al. [18] proposed a reaction product obstruct wetting theory for a Sn-0.3Ag-0.7Cu/Al reactive wetting system, and indicated that the reaction products were unfavorable to interfacial mass transfer and inhibited wetting. Satoshi et al. [19] reported the wetting behaviour of molten brazing filler metallic alloys on metallic substrate: the production of intermetallic compounds almost had not effect on wettability. In conclusion, interface will be inevitably produce intermetallic compound in the reactive wetting of metal/metal system. However, it is still controversial and there is no unified conclusion that the intermetallic compound and morphology have effect on the wettability. Meanwhile, these researchers indicate that wettability is associated with temperature and intrinsic properties of liquid metals, they also cannot establish a relationship between wettability and the interfacial mass transfer. Besides, the effect of Mg, Si elements and interfacial microstructure evolution remains obscure. For these reasons, the wetting and interfacial microstructure evolution warrant further study.

In this study, re-melt 6061 and 4043 alloys were selected: 6061 and 4043 are filler materials used when joining Al and Ti alloys. The spreading dynamics, interfacial microstructures and wettability of these two alloys on the pure Ti substrate were studied. To further reveal the intrinsic mechanism of interfacial compound evolution, the microstructures of 6061 and 4043 Al/Ti interface were analyzed. Besides, the spreading dynamic models were adopted to reveal the limited factors of wetting in the two systems.

2. Experimental procedure

TA2 pure titanium was selected as the base metal material with dimensions of 20 × 20 × 2 mm, 6061 and 4043 filler wire were re-melted to cubes with 2 × 2 × 2 mm for the later testing. The nominal chemical compositions of pure Ti, 6061 and 4043 alloys are summarised in Table 1. Prior to the experiment, the oxide film on the surface of Al alloy cubes were cleaned using an automatic polishing machine. The average roughness of the polished TA2 substrates were 40 nm, as measured by DEKTAK 6 M (Veeco Metrology Corp., Woodbury, NY, USA) over a distance of 2 mm at a speed of 100 nm/s. Then the TA2 substrate, 6061 and 4043 alloys cube block were immersed in a beaker of alcohol and ultrasonically cleaned for 10 min.

A modified sessile drop method [20] (when plate is heated to the desired experimental temperature, a magnetic push-rod moves the metal to the surface of the plate) was used to study wettability of an Al/Ti system at high temperatures, which can avoid the reaction between the droplet and plate in the heating process compared to the traditional sessile drop method. The wetting experiment was performed using an advanced LWAT/H-1 wetting system, and the measurement accuracy of contact angles reached ±0.1 degree. Prior to the wetting experiment, TA2 substrate was placed at the horizontal ceramic heating zone in the chamber of a wetting furnace. The chamber was evacuated to a high vacuum state, and the chamber was heated at a rate of 20 K/min to the experimental temperature. When the experimental temperature was stable, an Al cube was dropped onto the surface of Ti substrate. During the experiment, the clear profiles of the molten drop were obtained by CCD camera (at a laser backlight wavelength of 650 nm).
After the experiment, ultra-high-purity argon atmosphere (99.999%) was pumped into the chamber as a shielding gas until the specimens were cooled to 303 K. The wetting samples were cross-sectioned perpendicular to the radial direction for metallographic analysis. Backscattered electron image observation and quantitative compositions identification of the interfacial microstructure were conducted by scanning electron microscope (SEM, MERLIN Compact) equipped with an energy-dispersive X-ray spectrometer (EDS). The phases were identified by X-ray diffraction (XRD, D8 Advance).

3. **Results**

3.1. **Wetting phenomena**

**Fig. 1** shows the variation of contact angles and normalized contact radii \( R/R_0 \) (where \( R \) is the dynamic contact radius, and \( R_0 \) is initial contact radius) with time for the molten Al alloys on the pure Ti plate. The contact angles decreased with time from 46 ± 0.1° for 6061Al and 121 ± 0.1° for 4043Al to stable equilibrium contact angles. Liquid 4043Al alloy was non-wetting the Ti plate at 873 K, while the 6061 Al alloy had excellent wettability (Fig. 1(a)). \( R/R_0 \) varied with time monotonically, and there were two stages to the spreading process of the whole Al alloys (Fig. 1(b)). Non-linear spreading was presented in the first stage, and then the near-linear spreading was observed. The variation of \( R/R_0 \) to the non-linear spreading can be fitted by \( R/R_0 = R/R_0 - m \exp\left(-t/k\right) \), where \( R_0 \) is initial contact radius, \( m \) and \( k \) are fitting parameters [21]. The precursor film was produced in a near-linear stage, and the spreading rate in a 6061Al/Ti system was much better than a 4043Al/Ti system. The different initial wettability and spreading dynamics in these two systems caused by the Mg and Si elements in Al alloy.

3.2. **Macro-morphologies**

The macro-morphologies of the 6061 and 4043 alloys at pure Ti plates were shown in Fig. 2. 6061Al alloys exhibited good wettablity and the whole substrate was covered by the precursor film at 973 K (Fig. 2(c)). At the same experimental temperature, the final wettability for 6061 alloy was much better than 4043 alloy, and the width of precursor film was larger. At the triple line region, the precursor film clearly showed two layers. The extended rate of precursor film was much faster than the movement of triple line, and then the Al droplet spreading on the surface of precursor film (reaction layer, i.e. inert surface) rather than the pure Ti. Therefore, the spreading dynamics was different from the “chemical reaction control” model in near-linear stage.

<table>
<thead>
<tr>
<th>Material</th>
<th>O</th>
<th>Fe</th>
<th>Si</th>
<th>Mn</th>
<th>Mg</th>
<th>Zn</th>
<th>Ti</th>
<th>Cu</th>
<th>Al</th>
</tr>
</thead>
<tbody>
<tr>
<td>6061 Al</td>
<td>–</td>
<td>0.70</td>
<td>0.60</td>
<td>0.15</td>
<td>0.90</td>
<td>0.25</td>
<td>0.15</td>
<td>0.15</td>
<td>Balance</td>
</tr>
<tr>
<td>4043 Al</td>
<td>–</td>
<td>0.80</td>
<td>5.00</td>
<td>0.05</td>
<td>0.05</td>
<td>0.10</td>
<td>–</td>
<td>–</td>
<td>0.305 Balance</td>
</tr>
<tr>
<td>TA2 Ti</td>
<td>0.01</td>
<td>0.30</td>
<td>0.15</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>Balance</td>
<td>–</td>
<td>–</td>
</tr>
</tbody>
</table>

### 3.3. Microstructures

**Fig. 3** shows the cross-sectional view and top-view of the microstructures at the triple line region, and the interfacial microstructure at the central position for 6061Al/Ti after isothermal wetting at 873 K. Precursor film exceeded the triple line with respective characteristics can be observed (Fig. 3(a)). The spreading rate of the precursor film was much faster than a molten droplet, and two different morphologies could be observed: the under-layer produced a continuous intermetallic compound layer, and the top-layer was residual liquid Al. Actually, the precursor film was the extended reaction layer. To further observe the morphology of the precursor film, top-view of triple line were selected, as the details showed in Fig. 3(c). Some acicular phase was segregated at the surface of precursor film after solidification. The spreading rate of precursor film for under-layer was slightly higher than a top-layer (Fig. 3(d)). Seen in combination with EDS and elemental surface distribution analysis, the under-layer was mainly consisted of Al3Ti, and the top-layer was residual liquid Al (Fig. 3(e)-(j)). The cross-sectional view of the interfacial structure at the central position was selected, as be showed in Fig. 3(b), and the continuous reaction layer beneath the drop can be observed. Some granular phase evenly dispersed into the droplet, points EDS analysis shown that the chemical composition of the continuous reaction layer was Al3Ti, and small amount of granular Al3Ti was distributed in the droplet near the interface zone. The results were showed in Fig. 3(b)-(j), which is corresponding to the positions of yellow and red crosses in Fig. 3(b). It was noteworthy that Mg was not found either in the precursor film or in the interface region, while Si segregation in the reaction layer was obvious. The concentration of Si far exceeded the original value (0.6 wt. % in Table 1) at reaction layer. Also, the element line distribution suggested the Si segregation in the reaction layer but no Mg track (Fig. 3(b)).

**Fig. 4** shows the cross-sectional view and top-view microstructures for 4043Al/Ti interface and triple line region after isothermal wetting at 873 K and 925 K. At 873 K, the reaction layer was continuous, and denser in Ti side (bottom layer) with thickness of 3 μm, the virgate and granular phase were uniform distribution in the upper layer near the droplet (Fig. 4(a)-(b)). At 925 K, the bottom layer and virgate phase disappeared at central position, while a small amount of vibrate phase also remained in the continuous interface layer near the triple line region (Fig. 4(c)-(d)). Based on the element line distribution and point analysis by EDS in Fig. 4(b)-(d) and (i)-(k), the colors in Fig. 4(i)-(k) were corresponding to the color of crosses in Fig. 4(b)-(d), suggested that granular phase was Al3Ti with Si-rich, virgate phase and bottom layer were the Ti5Al3Si12, the concentration of which was surprisingly high in the reaction layer almost 8–9 times than the original concentration in 4043.
alloy which suggested the Si was an interfacial active element. The precursor film also extended reaction layer with some residual liquid Al, the reaction layer was not so compact that some residual liquid Al permeated into the layer (Fig. 4(e)-(h)). The precursor film also had two layers, the bottom layer was Al₃Ti and the top layer was residual liquid Al with some granular phase, elemental surface distribution clearly showed the two layers (Fig. 4(h)). The wetting temperatures were marked by the red lines in Fig. 5. The solubility of Ti in Al was limited by the experimental temperature. Therefore, the intermetallic compound could quickly produce, and then further resisted interfacial reaction.

**Fig. 1** - The variations with time in the (a) dynamic contact angles and (b) normalized contact radius $R/R_0$ for molten 6061 and 4043 alloys on pure Ti surface at 873 K, 923 K, and 973 K.

**Fig. 2** - Macro-morphologies after isothermal wetting at different temperatures, (a)-(c) 6061Al/Ti and (d)-(f) 4043Al/Ti at 873 K, 923 K and 973 K, respectively.

**Fig. 6** shows the macro-morphologies and SEM images after corrosion (the solidified Al was removed by 0.5 mol/L sodium hydroxide aqueous solution) for 6061 and 4043Al/Ti samples at 923 K. The microstructures of interfacial production for 6061Al/Ti samples were different from that 4043Al/Ti, and the microstructures for beneath the drop were different from that at close to the triple line. A lot of loose granular phase and residual Al at beneath the 6061 alloy drop, while the granular phase were smaller and denser at triple line region, besides the virgate phase disappeared in all range of solid-liquid interface (Fig. 6(b)-(c)). At beneath the 4043 alloy drop, the interface distributed regular granular phase and the size was about 3
times than that of 6061 alloy sample; the triple line region also distributed dense granular phase with small amount of acicular phase, as shown in Fig. 6(b)-(c). By comparing to the XRD spectrum of original Ti surface, the main phases in reaction layer were Al₃Ti and some residual Al, and the precursor film was constituted of Al₃Ti and residual liquid Al for 6061Al/Ti sample. However, the main phase in the precursor film were Al₃Ti and some Ti₇Al₃Si₁₂ (ε₁) for 4043Al/Ti sample, were showed in Fig. 7 (corresponding from a to d, respectively), which were in according with the EDS results.

4. Discussion

4.1. Wetting mechanism and spreading dynamics for nonlinear stage

In this Al/Ti reactive wetting system, the wettability and spreadability are determined by many factors, such as the presence of an oxide film, interfacial reaction, a small amount of solute elements, etc. The oxide film as one of main factors, the polished surface of Ti plate was covered by the TiO₂ oxide film of about 2 nm [22], blocking triple line movement. In this 6061Al/Ti system, Mg vapor which around the drop can reduce the thickness of this TiO₂ oxide film according to thermodynamic data (2Mg(g)+TiO₂(s)=2MgO(s)+Ti(s), Δ_rG_m^° = -485300 + 208.3T, J mol⁻¹), exposing the Ti atoms to the substrate surface, thus increasing the surface energy of Ti substrate at triple line region, then the liquid Al would infiltrate the crack of oxide film and formation a precursor film, triggering the triple line movement and promote interfacial reaction. Besides, the formation of loose phase (Al₃Ti) at triple line region can also remove the oxide film, and then improve wettability. In 4043Al/Ti system, 4043 alloy had weak wettability on the pure Ti surface at 873 K, but the interface produced intermetallic compound with the continuous compact Ti₇Al₃Si₁₂ layer in the bottom (Fig. 4). The solubility of Ti in
liquid Al is rather small and the continuous compact reaction layer (Ti₇Al₅Si₁₂) would block the mass transfer of S/L interface at triple line region, and then restrain wetting and spread. The primary reaction product Ti₇Al₅Si₁₂ decomposed at 852 K [23]. However, high concentration of Si inhibited the decomposition of Ti₇Al₅Si₁₂ at 873 K. Once the continuous compact Ti₇Al₅Si₁₂ layer was formed, the mass transfer at close triple line would further slowdown, and deteriorate wettability. On the contrary, 4043 alloy had a good wettability on the surface of Ti plate at 923 K, the bottom layer and virgate phase had disappeared, the loosened layer was consisted of the granular Al₃Ti phase. The decomposition of Ti₇Al₅Si₁₂ and production of Al₃Ti (loosened structure) removed the oxide film and promoted mass transfer at triple line region, improving the wettability. Based on the above considerations, the formation of Ti₇Al₅Si₁₂ at L/S interface, as a compact intermetallic layer block the mass transfer and deteriorate wettability. The formation of a loose phase (Al₃Ti) at triple line zone would remove the oxide film and promote wettability.

As mentioned above that Si segregation at L/S interface can be found but no Mg track, the active element segregation at L/S interface is a prerequisite for the precipitation of reaction product when the concentration of the active elements is below the saturation precipitation concentration. Therefore, adsorption energy of Si and Mg at Al-Ti interface can be used for the prediction of Al-Ti interfacial structure before experiment, which can be using the following equations [24,25],

\[
E_{\text{Si(Al)}\rightarrow \text{Mg(Al)}} = E_{\text{Si}} \cdot \lambda_{\text{TiSi}}\cdot \lambda_{\text{AlSi}}\cdot \lambda_{\text{TiAl}}
\]

\[
E_{\text{Si(Al)}\rightarrow \text{Mg(Al)}} = E_{\text{Si}} \cdot \lambda_{\text{TiMg}}\cdot \lambda_{\text{AlMg}}\cdot \lambda_{\text{TiAl}}
\]
where \( \lambda_{ij} = (\Delta H_{ij}^{\text{ad}} + \Delta H_{ij}^{\text{ex}})/2 \) (3)

where \( E_{\text{Si}}^{\text{ad}} \) and \( E_{\text{Mg}}^{\text{ad}} \) are adsorption energy of Si and Mg at Al-Ti interface, \( m_i \) is a positive interfacial structure factor, \( \lambda_{ij} \) is the mole exchange energy for \( i \) and \( j \), \( \Delta H_{ij}^{\text{ad}} \) and \( \Delta H_{ij}^{\text{ex}} \) represent the mixing enthalpy of \( i \) and \( j \) in \( i \). The more negative the value of adsorption energy for Si and Mg, which is more inclined to segregate at Al-Ti interface. \( E_{\text{Si}}^{\text{ad}}/m \) and \( E_{\text{Mg}}^{\text{ad}}/m \) are -73.4 kJ/mol and 221.4 kJ/mol for Al-Si/Ti system and Al-Mg/Ti system, respectively. Therefore, Si is segregated at L/S interface.

In nonlinear stage, the IMCs were produced at L/S interface, and the reaction mechanism influence the spreading dynamics. The spreading dynamics of this system have typical the characteristics of reaction product control (RPC) limited model [17,24]. The wetting activation energy can be obtained based on RPC fitting equation,

\[
\cos \theta_S - \cos \theta = (\cos \theta_S - \cos \theta_0) \exp (-k_1 t)
\]

where the \( \theta_S \), \( \theta \), and \( \theta_0 \) are the equilibrium, dynamic and initial contact angles, respectively. \( k_1 \) is a kinetic constant, and can be deduced from the fitting results based on the variations of \( \theta \) with \( t \) in Fig. 1(a). The wetting activation energy of 6061 and 4043Al/Ti systems was further deduced from Arrhenius plots of \( k_1 \) and 1/\( T \) (Fig. 8), with values of 56 kJ/mol and 71 kJ/mol. For a 6061Al/Ti system, the activation energy (56 kJ/mol) is associate with the Al3Ti compound formation (30 kJ/mol) and the surface energies in difference per molar area between \( \alpha \)-Ti and TiO2 (27.2 kJ/mol). The per molar area surface energies are the 1650 mJ/m² [26] and 850 mJ/m² [27] for Ti and TiO2, respectively, molar surface area is 33975.4 m²/mol for \( \alpha \)-Ti. Further, we note that the Al3Ti compound forma-
tion can reduce the TiO₂ oxide film, which may be why the values of the wetting activation energy was slightly smaller. Therefore, the removal of the oxide film and interfacial reaction mainly contributed mainly to the spreading forces. For a 4043Al/Ti system, Ti₃AlSi₁₂ decomposition is key factor to promote wetting. Besides, production of Al₃Ti (loosened structure) removed the oxide film and promoted mass transfer at triple line region, improving the wettability, therefore the activation energy (71 kJ/mol) is corresponding to the Ti₃AlSi₁₂ decomposition energy (48 kJ/mol) and the surface energies in difference per molar area between α-Ti and TiO₂ (27.2 kJ/mol).

4.2. spreading dynamics for near-linear stage

Significantly, the precursor film occurred at the end of non-linear stage and the spreading rate of the precursor film was much greater than the droplet. Besides, the precursor film was consisted of the granular phase Al₃Ti and residual liquid Al at the surface of former (Fig.7), proved that the continuous granular phase prevents the mass transfer. When the precursor film precipitated, actually the droplet further spread on the precursor film rather than pure Ti surface. The spreading dynamic may be different from the RPC model at triple line region in nonlinear stage, and the interfacial reaction is weak. Therefore, inert forces should be considered as the factor of limited spreading. In order to confirm the limited factors of control the droplet spreading, some other factors should be taken into consideration, such as adsorption/desorption in the triple line region, density, viscosity, surface/interfacial tension etc., and then three typical inert wetting dynamic models are introduced.

The adsorption/desorption of fluid elements in the triple line region maybe affect the droplet spreading for the molecular dynamic model. The relationship of $U$ (spreading rate) and $\theta$ (dynamic contact angle) as following [28,29],

$$U = 2\lambda K_s \sinh(\sigma_\nu (\cos \theta - \cos \theta) / 2n k_B T)$$

Where $K_s$ is fitting parameter, $k$ is distance between two adsorption or desorption sites, $K_s$ is the Boltzman constant, $\sigma_\nu$ is $L/V$ surface energy. The spreading rate $U$ as showed in Fig. 2. The fitting of $U$ and $\theta$ to this model shows a poor fitness, with $R$ square are the 0.479-0.581, 0.427-0.615 for 6061 and 4043 at 873K–973 K, respectively. Therefore, adsorption/desorption of fluid elements in the triple line region is not the limiting factor affecting spreading in this system.

Jiang’s empirical formula incorporate both the viscous dissipation-limited and molecular dynamic effects, the factor of affect spreading maybe associate with density, viscosity,

### Table 2 – Physical parameters used to fitting the dynamic models.

<table>
<thead>
<tr>
<th>T, K</th>
<th>$\sigma_\nu$, mN/m [26] 6061 Al</th>
<th>$\sigma_\nu$, mN/m [33] 4043 Al</th>
<th>$\mu$, mN·s/m² [26]</th>
<th>$\rho$, kg/m³ [26]</th>
</tr>
</thead>
<tbody>
<tr>
<td>873</td>
<td>925</td>
<td>912</td>
<td>1.41</td>
<td>2471</td>
</tr>
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<td>923</td>
<td>914</td>
<td>903</td>
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<tr>
<td>973</td>
<td>900</td>
<td>891</td>
<td>1.18</td>
<td>2245</td>
</tr>
</tbody>
</table>
surface/interfacial tension etc., and it can be expressed as following [30,31],

$$\frac{\cos \theta - \cos \theta_0}{\cos \theta + 1} = \tanh \left[ 4.96 \frac{\mu (\sigma / \eta)^{0.702}}{L} \right]$$ (7)

where \( \sigma_0 \) and \( \mu \) are the surface tension and liquid viscosity, \( \mu / \sigma_0 \) is a fitting parameter. The fitting parameter \( \mu / \sigma_0 \) are \( 1.61 \times 10^{-3} \), \( 1.49 \times 10^{-3} \) and \( 1.26 \times 10^{-3} \) for the 6061Al/Ti, at 873K, 923K and 973K, respectively. Physical parameters were shown in Table 2, substituting this value into the above fitting parameter, \( \mu / \sigma_0 \) can be calculated based on the physical parameters which are \( 1.5 \times 10^{-3} \), \( 1.4 \times 10^{-3} \) and \( 1.3 \times 10^{-3} \), respectively. Results show an excellent fitness for the 6061Al/Ti system, the fitting value is close to the actual value. However, 4043Al/Ti wetting systems shows a rather poor fitness (Fig. 9(b)).

In the hydrodynamic (namely, viscous dissipation-limited) model, the viscosity is the main factor of limiting the spreading. The relationship of dynamic contact angle \( \theta \) and spreading rate \( (L) \) as following [32],

\[
\theta^3 = 9(\mu / \sigma_0)\ln(L/L_0) \quad (8)
\]

\[
L = (2\sigma_0 / \rho g)^{1/2} \quad (9)
\]

where \( L \) and \( L_0 \) are the capillary length and slip length, respectively. \( \sigma_0, \mu, \rho, g \) are the surface tension, liquid viscosity, density and the gravitational constant, respectively. For convenience of fitting, we defined \( 9(\mu / \sigma_0)\ln(L/L_0) \) as a fitting parameter. The fitted results show an excellent fitness for the 4043Al/Ti wetting system, while the 6061Al/Ti wetting system show a poor fitness (Fig. 9(a)). The fitting parameters \( 9(\mu / \sigma_0)\ln(L/L_0) \) are \( 1.27 \times 10^5 \) and \( 1.18 \times 10^4 \) for the 4043Al/Ti at 923K and 973K, respectively. Physical parameters were shown in Table 2. \( L_0 \) can be calculated based on \( 9(\mu / \sigma_0)\ln(L/L_0) \) and in the order of \( 10^{-9} \) m. \( L_0 \) is the order of atomic or molecular dimension, \( 10^{-9}-10^{-8} \) implied the spreading was more inclined to the dependence of liquid viscous [32]. Therefore, in the near-linear stage, the spreading of 4043 alloy on precursor film was mainly determined by the viscous friction.

5. Conclusions

The wetting of pure Ti by Al alloys (6061 and 4043 Al) was investigated by modified sessile drop method at 873–973 K. The main conclusions are listed as follows:

(1) The wetting dynamic was determined by the reaction products at liquid/solid interface in the nonlinear stage. The formation of the Ti₅Al₃Si₂ at solid-liquid interface, as a compact intermetallic layer blocked the mass transfer and deteriorate wettability. The formation of a loose phase (Al₃Ti) at triple line zone could remove the oxide film and promote wettability.

(2) In nonlinear stage, the spreading dynamics can be described by RPC model, with the wetting activation energies are 56 kJ/mol and 71 kJ/mol for 6061 and 4043Al/Ti system. The activation energy (56 kJ/mol and 71 kJ/mol) was associated with the Al₅Ti compound formation (30 kJ/mol) or Ti₅Al₃Si₁₂ decomposition (48 kJ/mol) and the surface energies in difference per molar area between Ti and TiO₂ (27.2 kJ/mol).

(3) In near-linear stage, the droplets spread on the surface of intermetallic compounds. The spreading dynamic of 6061 alloy satisfies the Jiang’s empirical equation and 4043 alloy satisfies the viscous dissipation-limited model.

Conflicts of interest

There is no conflict of interest in this paper.

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