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Surface tension measurement of metastable liquid Ti–Al–Nb alloys

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Abstract Thermophysical properties of liquid alloys are usually difficult to measure, especially for high melting point and reactive alloys. In this work, the surface tensions of superheated and undercooled liquid $Ti_{55}Al_{45}$, $Ti_{50}Al_{45}Nb_5$ and $Ti_{45}Al_{45}Nb_{10}$ alloys are determined by using oscillating drop method under electromagnetic levitation state. The experimental results of Ti–Al and Ti–Al–Nb alloys display linear temperature dependence. The maximum undercoolings of 259 ($0.143T_L$), 268 ($0.146T_L$) and 275 K ($0.147T_L$) are respectively achieved for these three alloys. Furthermore, the viscosities of liquid $Ti_{55-x}Al_{45}Nb_x$ alloys are also derived from the experimental results.

1 Introduction

The thermophysical properties of liquid alloys are of both technical and scientific importance for liquid–solid phase transformation under nonequilibrium conditions [1–9]. As one of the most important thermophysical properties, surface tension is a key parameter for casting and welding processes, and also related to wetting phenomena [3]. Unfortunately, the surface tensions of undercooled liquids are still limited in the literature. The oscillating drop method together with electromagnetic levitation can provide a containerless environment and avoid the contamination of liquid surface. Therefore, this method is one of the most effective approaches for determining the surface tension of undercooled liquid alloys, especially for high melting point and reactive alloys [7, 8].

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Owing to their excellent applied properties, such as low density, high strength and resistance to oxidation, TiAl-based alloys are widely used as structural materials in aeronautics, astronautics and automotive industry [10–18]. Especially in recent years, Ti–Al–Nb alloys have aroused extensive research interest [13–16] because of their superior room-temperature ductility, higher yield stress, and enhanced oxidation resistance [18]. To perform quantitative research on the rapid solidification of Ti–Al–Nb alloys, their thermophysical properties are necessary.

In this work, the thermophysical properties of liquid $Ti_{55-x}Al_{45}Nb_x$ ($x = 0, 5, 10$) alloys are investigated. The objective is to determine the surface tensions and their temperature dependence at superheated and undercooled states by using the oscillating drop method. Furthermore, the viscosities of liquid $Ti_{55-x}Al_{45}Nb_x$ alloys are derived from the experimental results.

2 Method

$Ti_{55}Al_{45}$, $Ti_{50}Al_{45}Nb_5$ and $Ti_{45}Al_{45}Nb_{10}$ alloys were prepared from 99.999% purity Ti, 99.9999% purity Al and 99.99% purity Nb in an arc-melting furnace, and the mass of each sample was 0.5 g. Experiments were performed in an electromagnetic levitation facility, whose vacuum chamber was evacuated to 10^{-5} Pa and backfilled with argon gas. During the experiment, the levitated sample was melted by radio-frequency induction heating and then helium gas was blown toward the sample to achieve highly undercooled state. When an ideal undercooling was reached, the gas flow rate was properly regulated to preserve the liquid alloy at this undercooling for 10–30 s. The temperature was measured by an infrared pyrometer and the surface oscillations recorded by a photodetector at a sampling rate of 800 Hz.

The oscillation frequency was obtained by the fast Fourier transformation technique. Different spectra were derived at various temperatures and hence the temperature dependence of the surface tension could be obtained. The accuracy and validity of this experimental method have been verified and described in [8].

3 Results and discussion

By measuring the frequency of surface oscillations ω_R, the surface tension can be determined. Provided that the equilibrium shape of the droplet has a spherical shape and the oscillations are not damped, the surface tension σ for a droplet with mass m is related to ω_R by the Rayleigh equation [19]

$$\sigma = \frac{3}{8}\pi m\omega_R^2. \tag{1}$$

In practice, the levitated sample under terrestrial levitation conditions is usually distorted from its precisely spherical shape. As a result, this causes the oscillation frequencies to shift. Cummings and Blackburn considered the influence of a magnetic field on the split modes and derived the following frequency sum rule to obtain the Rayleigh frequency [20]

$$\omega_R^2 = \frac{1}{5}\sum_{l=-2}^{+2}\omega_{2,l}^2 - 2\varpi_{tr}^2, \tag{2}$$

where l is the split mode and ϖ_{tr} the mean translational frequency of droplet's center of mass. On the basis of (1) and (2), the Rayleigh frequency and surface tension can be derived from the frequencies of surface oscillations.

The surface tensions of liquid $Ti_{55}Al_{45}$, $Ti_{50}Al_{45}Nb_5$ and $Ti_{45}Al_{45}Nb_{10}$ alloys are determined over broad experimental temperature ranges of 1555–1954 K, 1569–2015 K and 1590–2044 K, and the maximum undercoolings of 259 ($0.143T_L$), 268 ($0.146T_L$) and 275 K ($0.147T_L$) are respectively achieved. Figure 1 presents the measured surface tensions of the three alloys. Linear relationships between surface tension and temperature can be described as follows

$$\sigma_{Ti_{55}Al_{45}} = 1.131 - 1.812\times10^{-4}(T-1814)\ \mathrm{N\,m^{-1}}, \tag{3}$$

$$\sigma_{Ti_{50}Al_{45}Nb_5} = 1.138 - 2.018\times10^{-4}(T-1837)\ \mathrm{N\,m^{-1}}, \tag{4}$$

$$\sigma_{Ti_{45}Al_{45}Nb_{10}} = 1.171 - 2.233\times10^{-4}(T-1865)\ \mathrm{N\,m^{-1}}. \tag{5}$$

According to (3)–(5), the surface tension values of liquid $Ti_{55}Al_{45}$, $Ti_{50}Al_{45}Nb_5$ and $Ti_{45}Al_{45}Nb_{10}$ alloys are determined to be 1.131, 1.138 and 1.171 $\mathrm{N\,m^{-1}}$ at their liquidus temperatures of 1814, 1837 and 1865 K, and their temperature coefficients are -1.812×10^{-4}, -2.018×10^{-4} and $-2.233\times10^{-4}\ \mathrm{N\,m^{-1}\,K^{-1}}$, respectively. With the increase

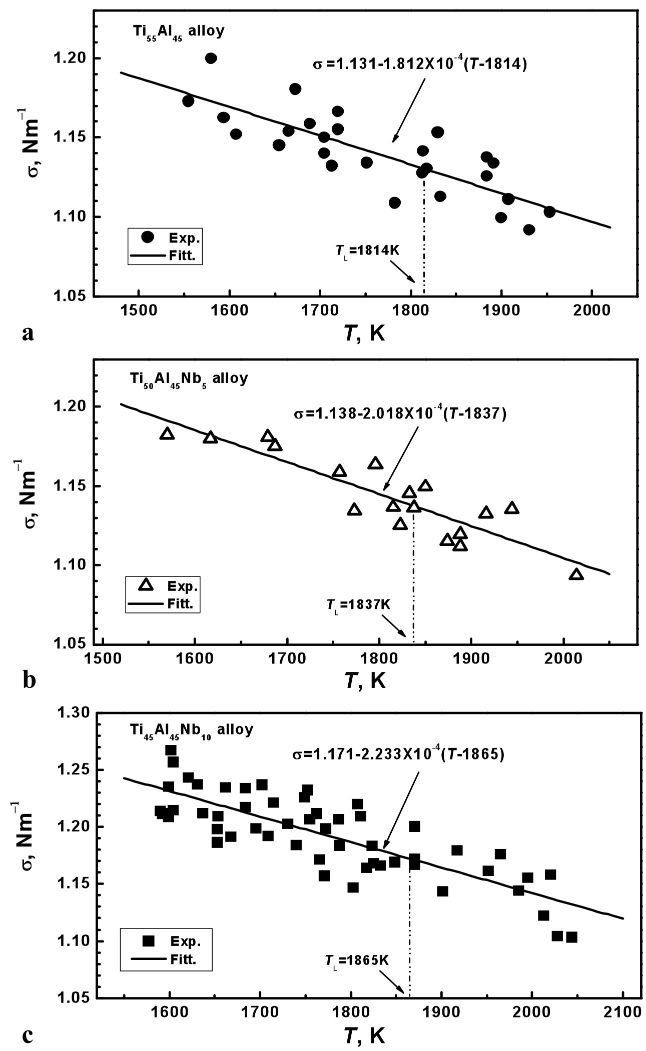

Fig. 1 Surface tension of liquid Ti–Al and Ti–Al–Nb alloys versus temperature: (**a**) $Ti_{55}Al_{45}$ alloy, (**b**) $Ti_{50}Al_{45}Nb_5$ alloy, and (**c**) $Ti_{45}Al_{45}Nb_{10}$ alloy

of niobium content, the surface tensions of Ti–Al–Nb alloys at liquidus temperature increase obviously.

For the sake of comparison, the experimental data of Ti, Al, Nb, Ti–Al, and Ti–Al–Nb alloys are listed in Table 1. The surface tension values of liquid $Ti_{55}Al_{45}$, $Ti_{50}Al_{45}Nb_5$ and $Ti_{45}Al_{45}Nb_{10}$ alloys are smaller than those of pure Ti [21] and Nb [21], but larger than that of pure Al [21] at liquidus temperature. In addition, when comparing the measured results of Ti–Al–Nb alloys with that of Ti–Al alloy [22], it is evident that the results change remarkably with the increase of niobium content. Moreover, when the compositions of niobium are close to each other, the difference among the surface tensions of $Ti_{50}Al_{45}Nb_5$, $Ti_{45}Al_{45}Nb_{10}$ and $Ti_{46}Al_{46}Nb_8$ [13–15] alloys is quite small.

The composition dependence of surface tensions for liquid ternary alloys is usually difficult to determine. On the basis of the experimental data, the surface tensions of liquid

Table 1 Surface tension values of liquid Ti, Al, Nb, Ti–Al and Ti–Al–Nb alloys

Materials	T_L (K)	σ_L ($\mathrm{N\,m^{-1}}$)	σ_T ($\mathrm{N\,m^{-1}\,K^{-1}}$)	T (K)	ΔT (K)	Reference
Ti	1958	1.650	-2.6×10^{-4}	1958	0	Iida [21]
Al	933	0.914	-3.5×10^{-4}	933	0	Iida [21]
Nb	2741	1.900	-2.4×10^{-4}	2741	0	Iida [21]
$Ti_{49}Al_{51}$	1753	1.094	-1.422×10^{-4}	1429–2040	324	Zhou [22]
$Ti_{55}Al_{45}$	1814	1.131	-1.812×10^{-4}	1555–1954	259	This work
$Ti_{50}Al_{45}Nb_5$	1837	1.138	-2.018×10^{-4}	1569–2015	268	This work
$Ti_{45}Al_{45}Nb_{10}$	1865	1.171	-2.233×10^{-4}	1590–2044	275	This work
$Ti_{46}Al_{46}Nb_8$	1842	1.114±0.05	$-(2.17\pm0.3)\times10^{-4}$	1613–1947	229	Nowak [13–15]

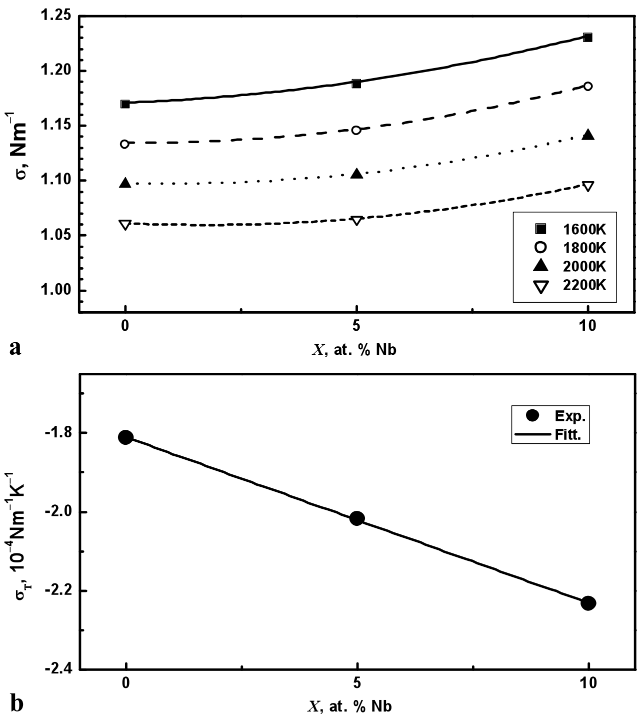

Fig. 2 Surface tension and temperature coefficient of liquid $Ti_{55-x}Al_{45}Nb_x$ alloys versus composition of niobium: (**a**) surface tension, (**b**) temperature coefficient

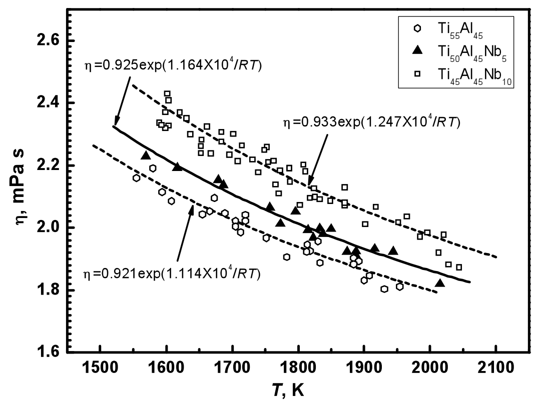

Fig. 3 Viscosity of liquid Ti–Al and Ti–Al–Nb alloys versus temperature

$Ti_{55-x}Al_{45}Nb_x$ alloys can be derived as shown in Fig. 2. All the magnitude of surface tensions of liquid $Ti_{55-x}Al_{45}Nb_x$ alloys increase with the increase of niobium content at different temperatures. When the temperature increases, the relevant surface tension decreases. In addition, their temperature coefficients decrease with the increase of niobium content.

The viscosity is also one of the important thermophysical properties of liquid alloys. Having obtained the surface tensions of undercooled liquid Ti–Al and Ti–Al–Nb alloys, the viscosity can be derived from the following theoretical model proposed by Egry [23]

$$\eta = \frac{16}{15}\sqrt{\frac{M}{kT}}\sigma, \tag{6}$$

where η is the viscosity, k the Boltzmann constant equal to $1.38\times10^{-23}\ \mathrm{J\,K^{-1}}$, and M the absolute atomic mass. According to (6), the viscosities of Ti–Al and Ti–Al–Nb alloys are obtained and presented in Fig. 3. The exponential regression of relevant data shows that viscosities are related to temperature as follows

$$\eta_{Ti_{55}Al_{45}} = 0.921\exp(1.114\times10^4/RT)\ \mathrm{mPa\,s}, \tag{7}$$

$$\eta_{Ti_{50}Al_{45}Nb_5} = 0.925\exp(1.164\times10^4/RT)\ \mathrm{mPa\,s}, \tag{8}$$

$$\eta_{Ti_{45}Al_{45}Nb_{10}} = 0.933\exp(1.247\times10^4/RT)\ \mathrm{mPa\,s}, \tag{9}$$

where R is the gas constant equal to $8.314\ \mathrm{J\,mol^{-1}K^{-1}}$. Based on (7)–(9), the viscosity constants of $Ti_{55}Al_{45}$, $Ti_{50}Al_{45}Nb_5$ and $Ti_{45}Al_{45}Nb_{10}$ alloys are equal to 0.921, 0.925 and 0.933 mPa s, respectively. The activation energies for viscous flow are 1.114×10^4, 1.164×10^4, and $1.247\times10^4\ \mathrm{J\,mol^{-1}}$. Obviously, the viscosities of liquid Ti–Al–Nb alloys increase with the content of niobium.

4 Conclusions

By using the oscillating drop method under electromagnetic levitation state, the surface tensions of superheated and undercooled liquid $Ti_{55}Al_{45}$, $Ti_{50}Al_{45}Nb_5$ and $Ti_{45}Al_{45}Nb_{10}$

alloys have been determined in the temperature ranges of 1555–1954 K, 1569–2015 K and 1590–2044 K. The samples are undercooled by up to 259 ($0.143T_L$), 268 ($0.146T_L$) and 275 K ($0.147T_L$), respectively. The experimental results exhibit linear temperature dependence. The viscosity values of undercooled liquid $Ti_{55}Al_{45}$, $Ti_{50}Al_{45}Nb_5$ and $Ti_{45}Al_{45}Nb_{10}$ alloys are also derived from the experimental results.

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