Solid-liquid Interface Energy of Metals at Melting Point and Undercooled State

Zengyun Jian[1], Kazuhiko Kuribayashi[1] and Wanqi Jie[2]

[1]*The Institute of Space and Astronautical Science, Sagamihara 229-8510, Japan*
[2]*State Key Laboratory of Solidification Processing, Northwestern Polytechnical University, Xi'an, 710072, P. R. China*

By investigating the effects of the configurational entropy, the vibrational entropy and the bonding strength of solid-liquid atoms on the structure of solid-liquid interface, a model for the interface energy of rough solid-liquid interface has been developed. From present model, the non-dimensional solid-liquid interface energies for metals at melting point are predicted to be 0.66–0.73, which are almost equal to the experimental result (0.66–0.75) obtained from grain boundary method. The solid-liquid interface energy decreases with increasing undercooling. At the maximum undercoolings that metals have reached, the non-dimensional solid-liquid interface energies predicted from present model are equal to 0.52–0.56. They are near to the experimental results (0.49–0.57) obtained from nucleation undercooling method. The predicted results of solid-liquid interface energy for metals from present model are in very good agreement with the experimentally measured results at melting point and undercooled state.

1. Introduction

The solid-liquid interface energy is an important physical parameter in nucleation and solidification theory. A complete comprehension of nucleation and solidification processing cannot be achieved without a clear knowledge of the solid-liquid interface energy.

Direct measurements of the solid-liquid interface energy have been possible in opaque alloy system[1–4] and transparent materials.[4–6] For opaque pure metals, it is difficult to directly measure the solid-liquid interface energy. Their values have usually had to be extrapolated from the dependence of the solid-liquid interface energy of alloy on composition, which can be measured by using grain boundary method,[3] or estimated from nucleation undercooling method on basis of the homogenous nucleation theory and the measured undercooling.[7–9]

Several theoretical models for the solid-liquid interface energy of metals have been made.[10–14] During these models, the most famous one is the model developed by Spaepen *et al.*[10, 11] In Spaepen model, the solid-liquid interface is supposed as a perfect smooth one. However, for metals, the solid-liquid interface is not a smooth one but a rough one. So, the predicted results from Spaepen model for metals are not only much higher the values from nucleation undercooling method for all metals but also are distinct higher than the results obtained from the grain boundary method for some metals. There are two models whose results are comparatively near to the values of solid-liquid interface energy measured from the grain boundary method and the nucleation undercooling method, respectively. One is the model developed by Grananfy *et al.*,[12] whose result is near to the experimental value of solid-liquid interface energy at undercooled state obtained from the nucleation undercooling method, but lower than the value at melting point measured from grain boundary. Another one is the model acquired by Jiang *et al.*,[14] the result of which comes near to the experimentally mea-

sured data at melting point from grain boundary method, but higher than that from nucleation undercooling method at undercooled state. So, at the moment, there is not a model of the solid-liquid interface energy for metals, whose results are not only in agreement with the measured values of solid-liquid interface energy at melting point, but also coincide to the experimental values obtained at undercooled state. So it is necessary to further investigate the model of the solid-liquid interface energy for metals.

The purpose of this paper is, by investigating the effects of thermodynamic elements on the structure of solid-liquid interface, to develop a model of the solid-liquid interface energy that matches the real structure of solid-liquid interface.

2. Solid-liquid Interface Energy of Metals at Melting Point

2.1 Solid-liquid interface energy of a perfect smooth interface

For convenience of investigation, we first discuss the solid-liquid interface energy in case that the interface is supposed as a perfect smooth one.

For condensed phases at ambient pressure, as shown in Fig. 1, the entropy in liquid phase S_l is greater than that in solid phase S_s. The difference of entropy between liquid phase and solid phase at melting point is the entropy of fusion ΔS_f.

The entropy of fusion consists two components: a vibrational part $\Delta S_{f,v}$ and a configurational part $\Delta S_{f,c}$. The vibrational part results from the increased local volume to the liquid atom for vibration around its average position. The configurational part is due to the number of configurations that are possible for a given energy of the random assembly of liquid atoms.

If the atoms in both crystal and liquid are thought to be vibrating about a fixed average position, for close packed metal, the difference in molar vibrational entropy between the two
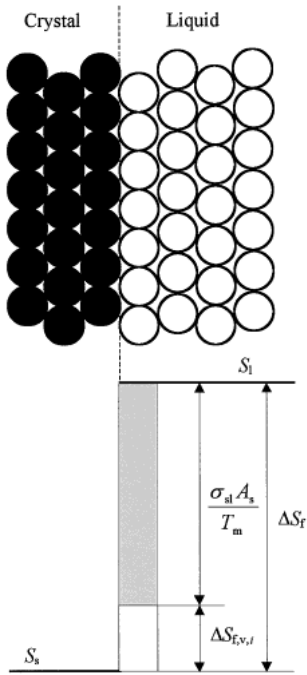
Fig. 1   Schematic representation of interface energy, entropy of fusion and the vibrational entropy of fusion in solid-liquid interface.

Table 1   Values of γ for some metals.[10]

Metals	γ
Silver	2.40
Copper	1.96
Nickel	2.01
Zinc	1.88

phases is determined by following equation:[10]

$$\Delta S_{f,v} = 3\gamma R \ln \frac{V_l}{V_s} \qquad (1)$$

where R is molar gas constant, V_s and V_l are the molar volumes of crystal and liquid respectively, γ is Grüneisen constant. The values of γ for some metals are shown in Table 1. It can be seen that the values of γ for metals are around 2.

Because the bounding strength for a bond of solid-liquid atoms is greater than that of liquid-liquid atoms, the first layer liquid atoms in the interface (*i.e.*, the liquid atoms neighbor directly with the solid atoms in the interface) cannot freely configure as the atoms in the bulk liquid. So the configuration of the liquid atoms in first layer of the interface should correspond to that of solid atoms. As is stated above, the configurational entropy depends on the configuration of atoms, so it can be thought that the configurational entropy of the liquid atoms in the first layer of interface should approximately be equal to that of the solid atoms. In other words, the configurational entropy of fusion for the liquid atoms in the first layer of interface should approximate to zero.

Therefore, if the vibrational entropy of fusion for the liquid atoms in first layer of interface is represented as $\Delta S_{f,v,i}$, for a perfect smooth interface as shown in Fig. 1, the relationship among $\Delta S_{f,v,i}$, ΔS_f and the solid-liquid interface energy σ_{sl} is

as follows:

$$\frac{\sigma_{sl} A_s}{T_m} = \Delta S_f - \Delta S_{f,v,i}, \qquad (2)$$

where A_s is the molar surface area of solid and it is in the form of:

$$A_s = b \sqrt[3]{V_s^2 N_A}, \qquad (3)$$

where N_A is Avogadro's constant, b is a coefficient depending on the structure of crystal. The value of b is equal to 1.09 for fcc and hcp crystals and it is equal to 1.12 for bcc crystal.[15]

Dividing eq. (2) by ΔS_f, we can obtain a non-dimensional solid-liquid interface energy ϕ_{sl}:

$$\phi_{sl} = \frac{\sigma_{sl} A_s}{T_m \Delta S_f} = \frac{\Delta S_f - \Delta S_{f,v,i}}{\Delta S_f}. \qquad (4)$$

Generally, the density of the atom in the solid-liquid interface should be greater than that of the atom in the bulk solid but lower than that of the atom in the bulk liquid. If it is supposed that the density of the atom in the solid-liquid interface is equal to the average of the densities of the bulk solid and liquid, the vibrational entropy of fusion for the liquid atoms in the first layer of interface, $\Delta S_{f,v,i}$, can be written as follows according to eq. (1):

$$\Delta S_{f,v,i} = 3\gamma R \ln \frac{V_l + V_s}{2V_s}. \qquad (5)$$

The calculated values of $\Delta S_{f,v,i}$ and ϕ_{sl} for some metals are shown in Table 2. It can be seen that the values of non-dimensional solid-liquid interface energy ϕ_{sl} for metals are in a range from 0.83 to 0.89 when the solid-liquid interface is supposed as a smooth one, which is similar to the result of the model obtained by Spaepen *et al.*[10,11] From their model, the non-dimensional solid-liquid interface energy is predicted to be 0.86 for fcc crystal on condition that the interface is thought as a perfect smooth one. But for metals, the solid-liquid interface is not a smooth one. So we must modify above model so as to make it match the structure of metals.

2.2 Modification on the perfect smooth model of solid-liquid interface

The equilibrium structure of solid-liquid interface should coincide to the state that the energy in the interface is at the minimum, which can be deduced in following procedures. When solid atoms are randomly added on a smooth solid-liquid interface, the interface will change in two aspects: producing new configurational entropy due to the random assembly of liquid and solid atoms ($S_{c,A}$), and producing new bonds of solid-liquid atoms in the interface layer (N_{sl}).

The produced configurational entropy $S_{c,A}$ can be calculated by following equation:

$$S_{c,A} = -kN[x \ln x + (1-x) \ln(1-x)], \qquad (6)$$

where N is the lattice numbers of atoms in the interface, k is Boltzmann's constant, x is the fraction of solid atoms in the interface.

The produced bonds of solid-liquid atoms in the interface layer N_{sl} can be written as follows:

$$N_{sl} = N Z_i x(1-x), \qquad (7)$$

Table 2 Predicted values of $\Delta S_{f,v,i}$ and ϕ_{sl} for some metals.

Elements	ΔS_f (Jmol^{-1} K^{-1})[7]	$V_s \times 10^6$ (m^{3}mol^{-1})[15]	$V_l \times 10^6$ (m^{3}mol^{-1})[15,16]	$\Delta S_{f,v,i}$ (Jmol^{-1} K^{-1})	ϕ_{sl}
Gold	9.56	10.76	11.39	1.44	0.85
Silver	9.12	11.16	11.54	1.01	0.85
Aluminum	11.45	10.59	11.41	1.89	0.83
Copper	9.59	7.61	7.91	1.03	0.89
Iron	8.86	7.66	7.98	1.03	0.88
Nickel	10.73	7.11	7.57	1.59	0.85
Cobalt	8.70	7.21	7.60	1.03	0.88
Platinum	9.62	9.68	10.15	1.20	0.88

where Z_i is the number of the nearest neighbors surrounding around an atom in the interface layer.

From eq. (4), we can obtain the energy for one bond of solid-liquid atoms ΔE_{sl}:

$$\Delta E_{sl} = \frac{\phi_{sl} T_m \Delta S_f}{N Z_n}, \qquad (8)$$

where Z_n is the number of the nearest neighbors in the next layer adjoining with an atom in the interface layer.

The product of ΔE_{sl} and N_{sl} is the produced energy due to producing new bonds of solid-liquid atoms in the interface layer $\Delta E_{sl,A}$:

$$\Delta E_{sl,A} = \frac{\phi_{sl} T_m \Delta S_f Z_i x(1-x)}{Z_n}. \qquad (9)$$

The total variation of energy due to adding solid atoms on a smooth solid-liquid interface $-\Delta G_{sl,A}$ is the sum of $-T_m \Delta S_{c,A}$ and $\Delta E_{sl,A}$. The value of N in eqs. (6), (7) and (9) is equal to Avogadro's constant N_A when the solid-liquid interface is in a molar area. So, for a molar area of interface, $\Delta G_{sl,A}$ can be written as follows:

$$\Delta G_{sl,A} = N_A k T_m [\alpha x(1-x) + x \ln x + (1-x) \ln(1-x)], \qquad (10)$$

where

$$\alpha = \frac{\phi_{sl} \Delta S_f \xi}{N_A k}, \qquad (11)$$

$$\xi = \frac{N_i}{N_n}. \qquad (12)$$

The values of ξ for fcc, bcc and hcp crystals are equal to 6/3, 4/2 and 6/3 respectively, in other words, they are equal to 2. Considering from the structure homogeneity of crystal, the values of ξ for other types of structure should be approximately 2 also. So we can think that ξ is equal to 2.

The equilibrium structure of solid-liquid interface should correspond to the state at which $\Delta G_{sl,A}$ is at the minimum. Analyzing eq. (10) mathematically, it is found $\Delta G_{sl,A}$ is at its minimum at the point that x is equal to 0.5 when α is not grater than 2, and the minimum of $\Delta G_{sl,A}$ is:

$$\Delta G_{sl,A}^* = N_A k T_m (0.25\alpha + \ln 0.5), \quad (\alpha \leq 2) \qquad (13)$$

where $\Delta G_{sl,A}^*$ is the minimum of $\Delta G_{sl,A}$. When α is greater than 2, the value of x corresponding to the minimum of $\Delta G_{sl,A}$ deviates from 0.5. Its value—x^* can be determined

by differentiating eq. (10) and letting it be zero:

$$\frac{\ln(1-x^*) - \ln x^*}{1 - 2x^*} = \frac{2\phi_{sl}\Delta S_f}{N_A k} = \alpha. \qquad (14)$$

The corresponding $\Delta G_{sl,A}^*$ is:

$$\Delta G_{sl,A}^* = N_A k T_m \lfloor \alpha x^*(1-x^*) + x^* \ln x^* + (1-x^*) \ln(1-x^*) \rfloor. \quad (\alpha > 2) \qquad (15)$$

If the solid-liquid interface energy corresponding to $\Delta G_{sl,A}^*$ is expressed as ϕ_{sl}^*, the relationship among ϕ_{sl}^*, ϕ_{sl} and $\Delta G_{sl,A}^*$ is:

$$\sigma_{sl}^* A_s = \sigma_{sl} A_s + \Delta G_{sl,A}^*. \qquad (16)$$

From eqs. (3)–(5), (13) and (15), we can obtain the equilibrium solid-liquid interface energy of metals:

$$\phi_{sl}^* = \beta \left(1 - \frac{6}{\Delta S_f} \ln \frac{V_s + V_l}{2V_s} \right), \qquad (17)$$

where

$$\phi_{sl}^* = \frac{\sigma_{sl}^* b^3 \sqrt{N_A V_s^2}}{T_m \Delta S_f}, \qquad (18)$$

$$\beta = 1.5 + \frac{2 \ln 0.5}{\alpha}, \quad (\alpha \leq 2) \qquad (19)$$

$$\beta = 1 + 2x^*(1-x^*) + \frac{2x^* \ln x^* + 2(1-x^*) \ln(1-x^*)}{\alpha}, \quad (\alpha > 2) \qquad (20)$$

The predicted values of α, β, ϕ_{sl}^* and σ_{sl}^* for some metals are shown in Table 3. It can be seen that the predicted values of non-dimensional solid-liquid interface energy for metals are in a region from 0.66 to 0.73.

3. Solid-liquid Interface Energy of Metals at Undercooled State.

For undercooled state, the solid-liquid interface energy of a perfect smooth interface can be expressed as an equation similar to eq. (2):

$$\frac{\sigma_{sl,T} A_{s,T}}{T} = \Delta S_{f,T} - \Delta S_{f,v,i,T}, \qquad (21)$$

where T is temperature, $\sigma_{sl,T}$, $A_{s,T}$, $\Delta S_{f,v,i,T}$, and $\Delta S_{f,T}$ are the solid-liquid interface energy, molar surface area of solid, the vibrational entropy of fusion in the interface and entropy of fusion at T, respectively.

Table 3   Predicted values of α, β, ϕ_{sl}^* and σ_{sl}^* for some metals.

Elements	α	β	ϕ_{sl}^*	σ_{sl}^* (Jm^{-2})
Gold	1.96	0.79	0.67	0.187
Silver	1.95	0.79	0.70	0.168
Aluminum	2.29	0.88	0.73	0.167
Copper	2.05	0.81	0.72	0.256
Iron	1.88	0.76	0.67	0.270
Nickel	2.19	0.85	0.72	0.382
Cobalt	1.84	0.75	0.66	0.293
Platinum	2.04	0.81	0.71	0.331

Table 4 $C_{p,s}$ and $C_{p,l}$ of metals.

Elements	$C_{p,l}$ (Jmol^{-1} K^{-1})[17]	$C_{p,s}$(Jmol^{-1} K^{-1})[18]
Gold	30.96	$23.66 + 5.18 \times 10^{-3}T$
Silver	30.56	$21.31 + 8.54 \times 10^{-3}T$ $+1.51 \times 10^5 T^{-2}$
Aluminum	31.80	$20.68 + 12.39 \times 10^{-3}T$
Copper	33.00	$22.65 + 5.86 \times 10^{-3}T$
Iron	43.05	$20.27 + 12.54 \times 10^{-3}T$
Nickel	39.30	$25.08 + 7.52 \times 10^{-3}T$
Cobalt	40.38	40.13
Platinum	36.50	$24.24 + 5.35 \times 10^{-3}T$

The values of $\frac{1}{V_T}\frac{dV_T}{dT}$ for solid and liquid metals are in a range from 10^{-5} to 10^{-6} K^{-1},[15, 16] where V_T is the molar volume of metal at T. Obviously, the effect of T on V_T is very trivial. So the values of $A_{s,T}$ and $\Delta S_{f,v,i,T}$ in eq. (21) can be thought to be approximately equal to their values at melting point, A_s and $\Delta S_{f,v,i}$, respectively.

$\Delta S_{f,T}$ depends on the specific heats of solid and liquid, $C_{p,s}$ and $C_{p,l}$, and undercooling ΔT (where $\Delta T = T_m - T$). The relationship among them is:

$$\Delta S_{f,T} = \Delta S_f + \Delta S_T, \tag{22}$$

where

$$\Delta S_T = \int_{T_m}^{T} \frac{C_{p,l} - C_{p,s}}{T} dT. \tag{23}$$

Practicing the subsequent procedures that we have done above in the investigation of the solid-liquid interface energy at melting point for metals to modify eq. (21), we can obtain the equilibrium solid-liquid interface energy of metals at undercooled state:

$$\phi_{sl,T}^* = \beta_T \left(1 - \frac{6}{\Delta S_f} \ln \frac{V_s + V_l}{2V_s} \right.$$
$$\left. + \frac{1}{\Delta S_f} \int_{T_m}^{T} \frac{C_{p,l} - C_{p,s}}{T} dT \right) \frac{T}{T_m}, \tag{24}$$

where

$$\phi_{sl,T}^* = \frac{\sigma_{sl,T}^* b^3 \sqrt{N_A V_s^2}}{T_m \Delta S_f}, \tag{25}$$

$$\beta_T = 1.5 + \frac{2\ln 0.5}{\alpha_T}, \quad (\alpha_T \leq 2) \tag{26}$$

$$\beta_T = 1 + 2x_T^* \left(1 - x_T^* \right)$$
$$+ \frac{2x_T^* \ln x_T^* + 2\left(1 - x_T^*\right)\ln\left(1 - x_T^*\right)}{\alpha_T}, \quad (\alpha_T > 2) \tag{27}$$

$$\alpha_T = \frac{2\phi_{sl}\Delta S_f}{N_A k} + \frac{2}{N_A k}\int_{T_m}^{T} \frac{C_{p,l} - C_{p,s}}{T} dT$$
$$= \frac{\ln\left(1 - x_T^*\right) - \ln x_T^*}{1 - 2x_T^*}, \tag{28}$$

where $\phi_{sl,T}^*$ and $\sigma_{sl,T}^*$ are the non-dimensional solid-liquid interface energy and solid-liquid interface energy of metals at

Table 5   Values of the maximum undercooling obtained in some metals.

Elements	T_m (K)	ΔT (K)	$\frac{\Delta T}{T_m}$
Gold	1336	230[7]	0.17
Silver	1234	260[9]	0.21
Aluminum	933	190[8]	0.20
Copper	1356	238[9]	0.18
Iron	1809	295[19]	0.16
Nickel	1726	365[19]	0.21
Cobalt	1768	330[19]	0.19
Platinum	2042	370[19]	0.18

Table 6   Predicted values of solid-liquid energy at the maximum undercooling listed in Table 5 from present model for some metals.

Elements	ΔS_T (Jmol^{-1} K^{-1})	α_T	β_T	$\phi_{sl,T}^*$	$\sigma_{sl,T}^*$ (Jm^{-2})
Gold	−0.19	1.91	0.77	0.53	0.144
Silver	0.06	1.96	0.79	0.56	0.135
Aluminum	−0.18	2.24	0.85	0.55	0.125
Copper	−0.60	1.90	0.77	0.53	0.189
Iron	−0.35	1.82	0.75	0.52	0.210
Nickel	−0.44	2.08	0.83	0.53	0.281
Cobalt	−0.04	1.83	0.74	0.53	0.235
Platinum	−0.46	1.93	0.78	0.53	0.247

undercooled state for a real solid-liquid interface.

In order to use above model to determine the values of solid-liquid interface energy of metals at undercooled state, we should know not only the undercooling, but also the specific heats of solid metal and undercooled liquid metal. The specific heat of solid metal varies with temperature. The relationship between the specific heat of solid metal and temperature can be directly measured by experiment. The specific heat of liquid metal is a constant for many metals.[17] The values of $C_{p,s}$, $C_{p,l}$ and the maximum undercoolings that metals have reached are listed in Table 4 and Table 5 respectively.

According the values of $C_{p,s}$ and $C_{p,l}$ in Table 4, and ΔT in Table 5, we have determined the solid-liquid interface energies for the metals at the undercoolings. The results are listed in Table 6. It can be seen that the values of non-dimensional solid-liquid interface energy for the metals at their maximum undercoolings are in a region from 0.52 to 0.56.

4. Comparison and Discussion

4.1 Comparison of present result for metal at melting point with the value of solid-liquid interface energy measured from grain boundary method

Miller and Chadwick[3] performed grain boundary melting-type experiments. From the measured curves of the solid-liquid interface energy of alloys as a function of the composition, they extrapolated the values of solid-liquid interface energy for pure metals at the melting point. It can be concluded that, from their experimental results, the non-dimensional solid-liquid interface energy for metals at melting point is in a region from 0.66 to 0.75. As is listed in Table 3, from present model, the predicted non-dimensional solid-liquid interface energies at melting point for metals are in a region from 0.66 to 0.73. Obviously, the solid-liquid interface energies from present model are in very good agreement with the experimental results obtained by Miller and Chadwick.[3]

Gündüz and Hunt[1,2] have developed a technique to routinely measure the solid-liquid interface energies in eutectic systems. By measuring the shape of grain boundary cusps after annealing in a temperature gradient, the solid-liquid interface energies and grain boundary energies for Al–Si, Al–Cu and Al–Mg alloys have been obtained. The results are listed in Table 7. On basis of their results, we can sketch the curve of the grain boundary energy (σ_{gb}^*) or the ratio of solid-liquid interface energy to the grain boundary energy ($\sigma_{sl}^*/\sigma_{gb}^*$) as a function of the solute content in aluminum alloys (c_w). The results are showed in Fig. 2. It can be seen that curve of $\sigma_{sl}^*/\sigma_{gb}^*$ or σ_{gb}^* as a function of c_w is a smooth straight line. So we can extrapolate the two smooth straight lines to the point at which c_w is zero to obtain the values of σ_{gb}^* and $\sigma_{sl}^*/\sigma_{gb}^*$ for pure aluminum. The obtained values of $\sigma_{sl}^*/\sigma_{gb}^*$ and σ_{gb}^* for pure aluminum are 0.5015 and 0.34 Jm^{-2}, respectively. Since the product of σ_{gb}^* and $\sigma_{sl}^*/\sigma_{gb}^*$ is σ_{sl}^*, it can be calculated that the value of solid-liquid interface energy (σ_{sl}^*) for pure aluminum is 0.1705 Jm^{-2}. The non-dimensional solid-liquid interface energy (ϕ_{sl}^*) corresponding to 0.1705 Jm^{-2} is calculated to be 0.7296, which is almost equal to the predicted result (0.73) in Table 3 from present model. So the solid-liquid interface energy for aluminum predicted from present model is in very good agreement with the experiment result obtained by Gündüz and Hunt too.

4.2 Comparison of present result with the experimental value of solid-liquid interface energy for metal at undercooled state obtained from nucleation undercooling method

On basis of homogenous nucleation theory, the solid-liquid interface energy of metal at undercooled state can be estimated from the measured undercooling. The accuracy of this method depends on whether the measured undercooling is that for homogenous nucleation. If it is not the undercooling for homogenous nucleation, the estimated result will be less than its real value. Table 8 is the obtained values of $\sigma_{sl,T}^*$ and $\phi_{sl,T}^*$ from the undercooling measured in metals on basis of homogeneous nucleation theory. The values of $\phi_{sl,T}^*$ from present model and the percent error of $\phi_{sl,T}^*$ between present model and the nucleation undercooling method are listed in Table 8 too. It can be seen that the results from present model are near to the experimental results obtained from the measured undercooling on basis of nucleation theory. The errors between present model and nucleation undercooling method for these metals are not greater than 8%.

For silver, nickel, copper, iron, cobalt and platinum, the values of solid-liquid interface energy from the nucleation theory method are equal to or almost equal to the results from present model, which indicates that the undercoolings obtained in these metals should have reached or almost have reached the values for them to nucleate homogeneously. Jian et al.[9] have proved that the nucleation form of silver with the undercooling in Table 5 is really a homogeneous one.

For gold and aluminum, the errors of solid-liquid interface energy between present model and the nucleation undercooling method are about 8%. The cause of producing these errors for aluminum and gold may be different. For gold, it may come from that only very limited experiments have been done on its undercooling because it is very expensive. The error for aluminum should result from its high chemical reactivity. As is well-known, aluminum is a very high chemical reactive el-
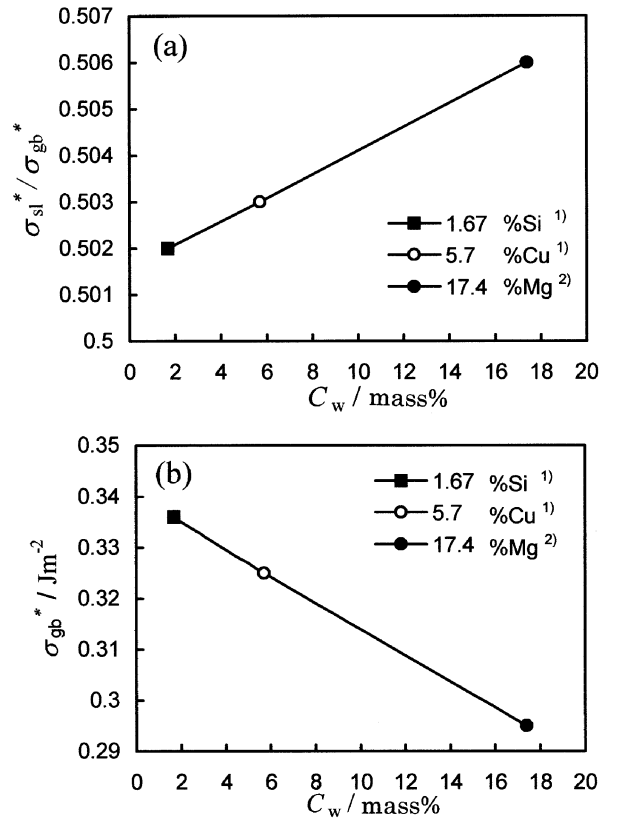

Fig. 2   The ratio of solid-liquid interface energy to grain boundary energy $\sigma_{sl}^*/\sigma_{gb}^*$ and the grain boundary energu σ_{gb}^* as a function of solute content c_w in aluminum alloys. (a) $\sigma_{sl}^*/\sigma_{gb}^*$, (b) σ_{gb}^*.

Table 7   The measured values of ϕ_{sl} and σ_{gb} in aluminum alloys.

Alloys	σ_{sl}^* (Jm^{-2})	σ_{gb}^* (Jm^{-2})	$\sigma_{sl}^*/\sigma_{gb}^*$
Al–1.67 mass%Si	0.1690[1]	0.336[1]	0.502
Al–5.7 mass%Cu	0.1634[1]	0.325[1]	0.503
Al–17.4 mass%Mg	0.1492[2]	0.295[2]	0.506

Table 8   Comparison of the present result with the experimental value of solid-liquid interface energy at the maximum undercoolings listed in Table 5 obtained from nucleation undercooling method.

Elements	$\sigma^*_{\text{sl},T}$ (Jm^{-2})	$\phi^*_{\text{sl},T}$	$\phi^*_{\text{sl},T}$	$\dfrac{\phi^*_{\text{sl},T}(\text{PM}) - \phi^*_{\text{sl},T}(\text{NU})}{\phi^*_{\text{sl},T}(\text{PM})}$
	Nucleation undercooling method (NU)		Present model(PM)	(%)
Gold	0.132	0.49	0.53	7.55
Silver	0.137	0.57	0.56	−1.79
Aluminum	0.116	0.51	0.55	7.27
Copper	0.185	0.52	0.53	1.89
Iron	0.204	0.51	0.52	1.92
Nickel	0.278	0.52	0.53	1.89
Cobalt	0.234	0.53	0.53	0
Platinum	0.240	0.52	0.53	1.89

ement. It is very difficult to obtain the state of homogeneous nucleation in aluminum melt because it is almost impossible to prevent aluminum react with oxygen and other elements.

4.3   Discussion

The solid-liquid interface of metals should be several atom layers in thickness in view of a large zone in the interface. But if we are in consideration of the solid-liquid interface in a small zone of atomic scale, it should be similar to present model in thickness or structure. So the results of solid-liquid interface energy for metals from present model not only are in very good agreement with the experimental results at melting point obtained from grain boundary methods, but also coincide to the values of solid-liquid interface energy at undercooled state attained from the measured undercoolings on basis of nucleation theory.

5.   Conclusions

A model for predicting the solid-liquid interface energy of metals at melting point and undercooled state has been developed. The solid-liquid interface energy for metals decreases with increasing undercooling. The predicted results of the solid-liquid interface energy from present model for metals are in very good agreement not only with the experimentally measured results at melting point from grain boundary method, but also with the experimental results at undercooled state obtained from nucleation undercooling method. In terms of present model, we can directly predict the solid-liquid interface energy of metals if knowing several physical parameters, which can be measured easily by experiments.

REFERENCES

1) M. Gündüz and J. D. Hunt: Acta Metall. **33** (1985) 1651–1672.
2) M. Gündüz and J. D. Hunt: Acta Metall. **37** (1988) 1839–1845.
3) W. A. Miller and G. A. Chadwick: Acta Metall. **15** (1967) 607–614.
4) S. C. Hardy: Philos. Mag. **35** (1977) 471–484.
5) J. G. Dash, V. A. Hodgkin and J. S. Wettlaufer: J. Statis. Phys. **95** (1999) 1311–1322.
6) L. A. Wilen and J. G. Dash: Science **270** (1995) 1184–1186.
7) D. Turnbull: J. Appl. Phys. **21** (1950) 1022–1027.
8) B. A. Mueller and J. H. Perepezko: Metall. Trans. **18A** (1987) 1143–1150.
9) Z. Y. Jian and Q. W. Jie: Metall. Trans. **31A** (2001) 559–563.
10) F. Spaepen: Acta Metall. **23** (1975) 729–743.
11) F. Spaepen and R. B. Meyer: Scipta Metall. **10** (1976) 257–263.
12) L. Grananfy, M. Tegze and A. Ludwig: Mater. Sci. Eng. **A133** (1991) 577–580.
13) F. Spaepen: Mater. Sci. Eng. **A178** (1994) 15–18.
14) Q. Jiang, H. X. Shi and M. Zhao: Acta Metall. **47** (1999) 2109–2113.
15) E. T. Turkdogen: *Physical Chemistry of High Temperature Techonlogy*, (Academic Press, New York, 1980) pp. 88–93.
16) R. L. David: *CRC Handbook of Chemistry of Chemistry and Physics*, (CRC Press, Tokyo, 1989) pp. B2–3, B215–220.
17) R. I. L. Guthrie and T. Iida: Mater. Sci. Eng. **A178** (1994) 35–41.
18) E. A. Brandes: *Smithells Metals Reference Book, 5th Edition*, (Butterworth & Co. Ltd., London & Boston, 1976) pp. 219–221.
19) M. C. Flemings and Y. Shiohara: Mater. Sci. Eng. **A65** (1984) 157–170.