Effects of Boron and Carbon on the Surface Tension of Molten Silicon under Precisely Controlled Oxygen Partial Pressure

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The surface tension of molten silicon is measured to be almost unchanged in the range of oxygen partial pressure, $P_{O_2} \leq 10^{-22}$ MPa with the sessile drop method under precisely controlled the oxygen partial pressure. Then, the effects of boron concentration (C_B /mass%) and carbon concentration (C_C /mass%) on the surface tension were investigated with the same method under $P_{O_2} \leq 10^{-22}$ MPa. The results obtained are summarized as follows. The surface tension increases with increasing C_B , and the maximum increase rate of the surface tension is about $30 \text{ mN} \cdot \text{m}^{-1} \cdot (\text{mass}\% C_B)^{-1}$ in the range of dilute boron concentration. The temperature coefficient of the surface tension, $(\partial\sigma/\partial T)_{C_B}$, increases with increasing C_B . Since dissolved boron from the BN substrate into molten silicon is below 0.054 mass% and the associated increase of the surface tension is below 1.5 mN/m, the contamination from the BN substrate on the surface tension can be ignored. The relation between the surface tension and C_B indicates negative adsorption and is well described with Szyszkowski's equation. The surface tension remains almost constant even in carbon saturated silicon and C_C has almost no influence on $(\partial\sigma/\partial T)_{C_C}$ in the range of $C_C \leq 84$ mass ppm ($C_{C,\text{sat}}$ at 1693 K).

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1. Introduction

Impurity distributions in silicon single crystals must be made as homogeneous as possible to fabricate ultra-large-scale-integrated circuits. The oxygen of around 10 mass ppm (10^{18} atoms/cm³) was dissociated from a quartz crucible into molten silicon and is transferred into the crystal through the melt convection in a Czochralski (CZ) method. Thus, the melt convection must be understood and controlled to obtain crystals with a homogeneous oxygen distribution. Several types of flow, such as natural forced and the surface tension induced (Marangoni) may be enhanced in CZ systems during crystal growth. Marangoni convection is thought to be a main cause of oxygen transfer into crystals. Therefore, understanding the flow, especially Marangoni flow that is originated by spatial distribution of the surface tension of molten silicon should be clarified.

In CZ crystal growth, the dopant such as B, P, As and Sb is already contained in the polycrystalline starting material or is added to the silicon melt. It is known that the oxygen concentrations in CZ silicon are affected by the doping elements.^{1,2} Carbon impurity in the silicon crystal produced by CZ method is introduced by carbon heaters or holders during CZ crystal growth via the silicon melts. The carbon impurity in silicon affects both oxygen precipitation and oxygen donor formation.^{3,4}

The authors^{5,6} have examined the effect of oxygen and temperature on the surface tension of molten silicon in the range of oxygen partial pressure, P_{O_2} , from 10^{-22} to 10^{-19} MPa by the sessile drop method. Relations between the temperature coefficient of the surface tension and P_{O_2} or the oxygen concentration in silicon (C_O /mass%) were fairly described quantitatively. The surface tension steeply decreases with increasing the P_{O_2} . While in the range of $P_{O_2} \leq 10^{-22}$ MPa, the change of the surface tension has a

tendency to becomes moderate. In order to detect precisely the effect of C_B and C_C on the surface tension, P_{O_2} in the atmosphere must be kept in an extremely low range where its influence on the surface tension is so small that the change in the surface tension due to the change in P_{O_2} during experiment can be neglected. Thus, this study confirmed the extent of P_{O_2} dependence of the surface tension in the condition of P_{O_2} lower than 10^{-22} MPa and then the effect of C_B and C_C on the surface tension was investigated under the P_{O_2} where the influence of P_{O_2} on the surface tension is neglected, that is, in the condition of P_{O_2} lower than 10^{-22} MPa.

2. Experimental Procedure

2.1 Apparatus

The experimental apparatus consisted of a LaCrO₃ heating element furnace, a gas purifier, oxygen sensor, a photographic system and a digital system (computer). In order to maintain air-tightness of the reaction chamber, a double tube structure was adopted in the study. The reaction tubes were made of high purity alumina (99.8% Al₂O₃). The outer tube is 50 mm in external diameter, 42 mm in internal diameter and 700 mm in length, and the inner tube is 37 mm in external diameter, 30 mm in internal diameter and 800 mm in length. Both ends of the reaction tube are sealed by water-cooled stainless caps. The temperature was measured with a 20:40Pt-Rh thermocouple, which was set directly under the boron nitride substrate. The maximum temperature for the furnace is 1923 K. The temperature was controlled by a PID digital program controller.

The droplet shape of molten silicon was taken using a camera connected with a telephotographic lens by a bellows. The focal distance must be constant in all the experiments to fix the magnification of the photographed image. The focal distance was present in the position of infinity and the focus was

Table 1 Chemical composition of CZ silicon and additions.

Sample	Si	O	B	C
CZ silicon	99.999 mass%	≈10 mass ppm	≤1 mass ppm	≤5 mass ppm
Boron lump	$B \geq 99.8 \text{ mass\%}$			
Graphite powder	Fixed carbon >99.6 mass%, Volatile <0.15 mass%, Ash <0.2 mass%			

adjusted by changing the distance between the molten droplet and the camera fixed on a mechanical table, which can be moved three dimensionally.

The argon gas was passed through an Ar gas purifier and further deoxidized by magnesium chips heated at around 823 K. The oxygen partial pressure in argon gas, P_{O_2} , was controlled below 10^{-22} MPa that is below $P_{O_2, \text{sat}}$ in equilibrium with $\text{SiO}_2(\text{s})$ by the aid of a gas deoxidized purifier and a tightly sealed furnace made of double alumina tubes. $P_{O_2}^{\text{out}}$, that is, P_{O_2} of Ar gas exhausted from the reaction chamber and $P_{O_2}^{\text{in}}$, that is, P_{O_2} of Ar gas before introducing into the heating furnace were measured with an oxygen sensor of $\text{ZrO}_2\text{--CaO}$ solid electrolytes using a three-way cock, respectively.

2.2 Procedure

CZ silicon was employed as the base liquid for all composition of materials prepared. The composition of CZ silicon single crystals and additions (boron and carbon) are shown in Table 1. The cylindrical CZ silicon (about 1.83 g) was polished with sandpaper to remove any surface oxide and cleaned with acetone using an ultrasonic automatic washer. The high purity boron nitride (99.5%BN) plate (25.7 mm × 25.7 mm × 2.5 mm) was used as a substrate in the experiment. The BN substrate was washed with ethanol, dried and used without touching the surfaces, in order to avoid any possible contamination.

As shown in Fig. 1, the silicon sample containing boron or carbon was set on the upper surface of the BN substrate, and the horizon of the substrate was adjusted by two water levels. After the sample was placed into the inner alumina reaction tube of furnace by alumina support, the system was sealed and evacuated by a vacuum pump. Then, introduction of argon gas into the reaction chamber and evacuated was repeated three times. After that, the system was heated to experimental temperature in an argon gas atmosphere. The total flow rate of argon was maintained at 0.16 L/min and the total pressure was held constant at 0.1 MPa in all the experiments.

The photographs of the droplet of molten silicon were taken every 5 min. The measurement time was 60 min for one experimental run. In order to obtain the values in the equilibrium state, the results measured in the latter 30 min were adopted.

According to Rotenberg's method,⁷⁾ the surface tension of molten silicon was calculated from the image contour of the droplet, which was input into a computer with a digitizer, by determining the curve with best fit between the numerical solution to the classical Laplace's equation and experimentally measured points. The magnification was determined by a picture of a standard steel ball (10 mm in diameter) taken with

Fig. 1 Schematic diagram of experimental apparatus.

the same camera and in the same focal distance. The density of Si-B ($C_B \leq 2.35 \text{ mass\%}$) system as a function of temperature and boron concentration can be written as follows:

$$\begin{aligned} \rho (\text{Mg} \cdot \text{m}^{-3}) = & (2.52 - 3.07 \times 10^{-2} [C_B / \text{mass\%}]) \\ & - (3.32 - 0.457 [C_B / \text{mass\%}]) \\ & + 0.105 [C_B / \text{mass\%}]^2 \times 10^{-4} (T - 1687) \end{aligned} \quad (1)$$

Equation (1) obtained by a modified sessile drop method⁸⁾ was used for determining the surface tension of the Si-B system. The density of molten silicon within saturated carbon concentration obtained in the study⁸⁾ is in agreement with the density of pure molten silicon. The measurement error was estimated to be $\pm 2\%$ in the surface tension and $\pm 0.5\%$ in the contact angle.⁹⁾

The boron concentration in silicon was analyzed by radiofrequency inductively coupled plasma (I.C.P.) emission spectrometry and carbon concentration in silicon by the combustion-IR absorption method.

3. Results and Discussion

3.1 Effect of P_{O_2} in argon gas

As shown in Fig. 2, the change of the surface tension with the oxygen partial pressure is very small in the range of

Fig. 2 Effect of the oxygen partial pressure in argon on the surface tension of molten silicon.

Fig. 3 Temperature coefficient of the surface tension of molten silicon as a function of oxygen partial pressure of argon gas.

$P_{O_2} \leq 10^{-22}$ MPa. The surface tension decreases remarkably with increasing the oxygen partial pressure in the range of P_{O_2} from 5×10^{-22} to 10^{-19} MPa, which is consistent with previous results obtained by the same apparatus and method.^{6,10)}

As shown in Fig. 3, the temperature coefficient of the surface tension, $K^P = (\partial\sigma/\partial T)_{P_{O_2}}$, is minus and increases with increasing the oxygen partial pressure. $(\partial\sigma/\partial T)_{P_{O_2}}$ at $P_{O_2} = 10^{-24}$ MPa is equal to about $-0.75 \text{ mN} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ and then $(\partial\sigma/\partial T)_{P_{O_2}}$ gradually increases with increasing the oxygen partial pressure up to 5×10^{-22} MPa. $(\partial\sigma/\partial T)_{P_{O_2}}$ steeply increases with increasing the oxygen partial pressure in argon gas in the range of P_{O_2} from 5×10^{-22} to 10^{-20} MPa.

$P_{O_2,\text{sat}}$ shown in Fig. 2 or Fig. 3 is the equilibrium oxygen partial pressure in $\text{SiO}_2(\text{s})\text{--Si(l)}\text{--O}_2$ system and can be calculated from $\Delta G^\circ = -952700 + 203.8T$ (J/mol).¹¹⁾

Fig. 4 Effect of boron concentration on the surface tension of Si-B system.

Fig. 5 Temperature dependence of the surface tension of molten silicon with various boron concentrations.

3.2 Effect of C_B on the surface tension

The surface tension of Si-B system was determined in the condition of $P_{O_2} \leq 10^{-22}$ MPa and C_B lower than the saturated boron concentration (about 3.2 mass%¹²⁾). Figure 4 shows results obtained in this study and those by other researchers.¹³⁾ The surface tension increases with increasing C_B , and the maximum increase rate of the surface tension in the range from 1693 to 1773 K is evaluated to be about $30 \text{ mN} \cdot \text{m}^{-1} \cdot (\text{mass}\%C_B)^{-1}$ from the slope of $\sigma - C_B$ relation at $C_B = 0$ (Fig. 4, 1773 K). Additionally, as temperature becomes higher, the increase rate of the surface tension with boron concentration is higher.

Figure 5 shows temperature dependence of the surface tension of Si-B system with various boron concentrations. The temperature coefficient of the surface tension, $K^B = (\partial\sigma/\partial T)_{C_B}$, is from -0.79 to $-0.35 \text{ mN} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ and in-

Fig. 6 Temperature coefficient of the surface tension as a function of boron concentration in silicon.

creases with increasing C_B as shown in Fig. 6. The decrease rate of the surface tension with temperature is lower as C_B becomes higher.

3.3 Equation of the surface tension and C_B

Szyszkowski¹⁴⁾ derived an empirical relation to account for the effect of surface-active elements on the surface tension of liquid iron in a binary melt. Belton¹⁵⁾ modified this equation and presented it in a more usable form, by combining the Gibbs adsorption isotherm with the Langmuir isotherm:

$$\sigma = \sigma^\circ - RT\Gamma_i^\circ \ln(1 + K_i a_i) \quad (2)$$

where σ is the surface tension (mN/m), σ° is the surface tension of the pure metal (mN/m), R is gas constant ($8.2056 \times 10^7 \text{ mN} \cdot \text{K}^{-1} \cdot \text{mol}^{-1}$), Γ_i° is the saturated surface excess concentration of the surface active component i (mol/cm²), K_i is the adsorption coefficient of component i and a_i is the activity of species i in solution.

Γ_i° can be determined by calculating the slope of the surface tension to the logarithm of the solute activity curve at high activities of solute according to the Gibbs adsorption equation.

$$\Gamma_i = -(1/RT)(d\sigma/d \ln a_i) \quad (3)$$

where Γ_i is the surface excess concentration (mol/cm²).

$d\sigma/d \ln a_i$ is minus ($\Gamma_i > 0$), which means positive adsorption; $d\sigma/d \ln a_i$ is plus ($\Gamma_i < 0$), which means negative adsorption. There have been a lot of reports relating to studies of positive adsorption. For example, Ogino *et al.*,¹⁶⁾ Takeuchi *et al.*¹⁷⁾ and Zhu *et al.*¹⁸⁾ have concluded that the influence of surface-active species such as Q and S on the surface tension of liquid iron is described with eqs. (2) and (3). But there have been little reports relating to negative adsorption. Monma and Suto¹⁹⁾ measured the surface tension of substitutionary binary alloy system, and concluded that the change of surface tension in a binary alloy system occurs in coincidence with the liquid line of phase diagram. This means that the surface tensions of the melt will increase when higher surface tension (higher

Fig. 7 Relation between the surface tension of Si-B system and $\ln(C_B/\text{mass}\%)$.

melting point) elements are added into lower surface tension (lower melting point) elements. This is in conformity with the results shown in Fig. 4.

The relation between the surface tension of Si-B system and $\ln(C_B/\text{mass}\%)$ is shown in Fig. 7. Since the each slope of the broken lines in the range of $C_B = 0.26 \sim 2.09 \text{ mass}\%$ in Fig. 7 shows almost constant, the saturated surface excess Γ_B° can be evaluated from the slopes as follows: -4.59×10^{-11} , -5.77×10^{-11} , -8.47×10^{-11} and $-1.05 \times 10^{-10} \text{ mol/cm}^2$ at 1693, 1703, 1713 and 1733 K, respectively. σ° was obtained by extrapolating the solid line in Fig. 4 to $C_B = 0$. Substituting Γ_B° , σ° , T and actual data into eq. (2), K_B can be calculated using the least square method.

Since the activity coefficient of boron at $X_B = 0.052$ ($C_B = 2.09 \text{ mass}\%$) is calculated to be 0.21, assuming the regular solution of Si-B alloy ($RT \ln \gamma_B = \alpha(1 - X_B)^2$) at 1693 K and using $\gamma_B^\circ = 0.18^{20)}$ (γ_B° is the activity coefficient of boron in silicon at $C_B \rightarrow 0$). f_B can be calculated to be 1.13 from $f_B = (\gamma_B/\gamma_B^\circ)(1 - X_B + X_B M_B/M_{Si})$ (M_B and M_{Si} are the atomic weight of boron and silicon, respectively). Therefore, the activity of boron in the range of $C_B \leq 2.09 \text{ mass}\%$ approximately obeys Henry's law ($a_B = f_B C_B \approx C_B$). Relation between the observed surface tension and boron concentration can be well described with eqs. (4)–(7).

$$\sigma = 821 + 6.37 \ln(1 + 4.49C_B), \quad 1693 \text{ K} \quad (4)$$

$$\sigma = 809 + 8.07 \ln(1 + 6.84C_B), \quad 1703 \text{ K} \quad (5)$$

$$\sigma = 799 + 11.9 \ln(1 + 4.40C_B), \quad 1713 \text{ K} \quad (6)$$

$$\sigma = 785 + 14.9 \ln(1 + 3.81C_B), \quad 1733 \text{ K} \quad (7)$$

The results calculated with eqs. (4)–(7) (the broken lines in Fig. 4) are in agreement with observed results.

The relation between C_B and the surface tension were measured in the condition of P_{O_2} from 1.62×10^{-25} to $2.36 \times 10^{-22} \text{ MPa}$. According to Fig. 2, it can be concluded

Fig. 8 Comparison of the measured values with the calculated values of the surface tension of Si-B-O system at 1693 K.

that the error of measuring data caused by the change of P_{O_2} during this experiment is smaller than 0.8%.

Equation (2) is available for describing the surface tension of Si-O system and is given by eq. (8).⁶⁾

$$\sigma = 845 - 29.7 \ln(1 + 6.62 \times 10^{10} P_{O_2}^{1/2}), \quad 1693 \text{ K} \quad (8)$$

Then, the surface tension of the present Si-O-B system may be described by simply combining eqs. (4) and (8), as shown with eq. (9), because the oxygen concentration is very small and the boron is not so strong surface active for molten silicon.

$$\sigma = 845 - 29.7 \ln(1 + 6.62 \times 10^{10} P_{O_2}^{1/2}) + 6.37 \ln(1 + 4.49 C_B) \quad 1693 \text{ K} \quad (9)$$

The values of surface tension of the experimental measurements and those calculated from eq. (9) are shown in Fig. 8. It can be seen that the values calculated from eq. (9) are the same or similar to the measured values. Ogino *et al.*¹⁶⁾ have reported that the similar treatment is also available for Fe-O-S system in the dilute concentration of oxygen and sulfur.

3.4 Contamination by the BN substrate

When using the sessile drop method to measure the surface tension, the main contamination is from the substrate directly contacting molten silicon.

In order to investigate the interfacial properties between molten silicon and solid materials and also to find a suitable substrate for the surface tension measurement with the sessile drop method, wettability and reactivity between molten silicon and various kind of plates were investigated under precisely controlled oxygen partial pressure. The oxide plates, such as SiO_2 , Al_2O_3 , MgO , CaO , ZrO_2 - Y_2O_3 , ZrO_2 - CaO and the carbide plate (SiC) considerably react with molten silicon and also show the good wettability, which indicates that those plates are unsuitable for the surface tension measurement. The BN plate is regarded to be the most suitable substrate because the BN has large contact angle ($145 \pm 5^\circ$) and shows low chemical reactivity with molten sil-

Fig. 9 Relation between temperature and boron concentration in silicon determined by ICP.

icon. The Si_3N_4 plate shows the least reactivity with molten silicon among the all kinds of materials investigated, but it has the contact angle about 90° , which causes large error in the surface tension measurement. Therefore, The BN was selected as the substrate for supporting the silicon drop in the present study.

It is very difficult to split the silicon drop after the measurement from the BN substrate at room temperature. After the measurement, the cross section of Si/BN interface was examined by an EPMA to know the reactivity at the interface of silicon and the BN substrate. It was confirmed that there was extreme trace of silicon in the BN side. However, in the silicon phase near the interface, it could be only confirmed the existence of N and it could not be confirmed the existence of boron. Therefore, it is inferred that a discontinuous Si_3N_4 layer is possibly formed at the interface, which restricts BN dissolving and spreading into molten silicon after the formation of the discontinuous Si_3N_4 layer.

Figure 9 shows C_B of the silicon drop after measurement and the results of molten silicon in the BN crucible after equilibrium experiment for 48 hours.²⁰⁾ C_B of CZ silicon sample (before measurement) was less than 1 mass ppm. After measurement, C_B in silicon drop without boron addition gradually increased as temperature increased. The maximum C_B dissolved from the BN substrate into molten silicon was 0.054 mass% in this study. The broken line in Fig. 9 indicates that even the reaction reaches equilibrium state, the maximum C_B at 1873 K is only 0.089 mass%. According to Fig. 4, the maximum increment of the surface tension is about $30 \text{ mN} \cdot \text{m}^{-1} \cdot (\text{mass}\% C_B)^{-1}$ in the range of dilute boron concentration. Hence the maximum surface tension increment by C_B dissolved from the BN substrate into molten silicon is below 1.5 mN/m, which shows that the influence by the BN substrate is much lower than the maximum error evaluated as $\pm 2\%$ for this measurement.

3.5 Effect of C_C on the surface tension

Figure 10 shows the relation between the surface tension of

Fig. 10 Effect of carbon concentration on the surface tension of molten silicon.

Fig. 12 Relation between temperature coefficient of the surface tension and carbon concentration in silicon.

Fig. 11 Phase diagram of the condensed phases at the Si-O-C system.

molten silicon and C_C in the range of P_{O_2} from 5.12×10^{-24} to 2.35×10^{-22} MPa. The surface tension of molten silicon remains almost constant even in carbon saturated silicon ($\log C_{C,sat} = 3.63 - 9660/T$, mass%).²¹⁾ Figure 11 shows phase diagram of the condensed phases at the Si-O-C system and is drawn from thermodynamic data.²²⁾ The point $P_{C,sat}$ at the co-existing line of Si(l) and SiC(s) is corresponding to the saturated carbon concentration in molten silicon ($C_{C,sat}$). When the partial pressure P_C is larger than or equal to $P_{C,sat}$, the silicon drop is in the condition that a SiC(s) film can be formed on the surface of molten silicon drop, which may cause the increase in the surface tension of molten silicon in the range of $P_C > P_{C,sat}$ as shown in Fig. 10. Figure 12 shows the relation between the temperature coefficient of the surface tension, $K^C = (\partial\sigma/\partial T)_{C_C}$, and carbon concentration in silicon. C_C has almost no influence on the temperature coefficient of the surface tension in the range of $C_C \leq 84$ mass ppm

($C_{C,sat}$ at 1693 K).

4. Conclusions

The effects of boron concentration (C_B /mass%) and carbon concentration (C_C /mass%) on the surface tension of molten silicon have been determined with the sessile drop method under precisely controlled the oxygen partial pressure in the range of $P_{O_2} \leq 10^{-22}$ MPa.

(1) The change of surface tension of molten silicon is very small in the range of $P_{O_2} \leq 10^{-22}$ MPa. The temperature coefficient of the surface tension, $(\partial\sigma/\partial T)_{P_{O_2}}$, increases with increasing P_{O_2} . $(\partial\sigma/\partial T)_{P_{O_2}}$ is equal to about $-0.75 \text{ mN}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ at $P_{O_2} = 10^{-24}$ MPa and then $(\partial\sigma/\partial T)_{P_{O_2}}$ gradually increases with increasing the oxygen partial pressure up to 5×10^{-22} MPa. $(\partial\sigma/\partial T)_{P_{O_2}}$ steeply increases with increasing the oxygen partial pressure in argon gas in the range of P_{O_2} from 5×10^{-22} to 10^{-20} MPa.

(2) The surface tension increases with increasing C_B , which shows negative adsorption of boron, and the maximum increase rate of the surface tension is about $30 \text{ mN}\cdot\text{m}^{-1}\cdot(\text{mass}\%C_B)^{-1}$ in the range of dilute boron concentration. The relation between the surface tension and C_B is fairly well described with Szyszkowski's equation. $(\partial\sigma/\partial T)_{C_B}$ also increases with increasing boron concentration.

(3) Since dissolved boron from the BN substrate into molten silicon is below 0.054 mass% and the associated increase of the surface tension is below 1.5 mN/m, the contamination from the BN substrate on the surface tension can be ignored.

(4) The surface tension of molten silicon remains almost constant even in carbon saturated silicon and C_C has almost no influence on $(\partial\sigma/\partial T)_{C_C}$ in the range of $C_C \leq 84$ mass ppm ($C_{C,sat}$ at 1693 K).

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