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密閉型ガスジェット浮遊装置による Hypercooling limit を利用した金属の融解熱測定

Measurement of the Heat of Fusion for Pure Metals using Hypercooling Limit with a Closed Type Aerodynamic Levitator

河本幹太 [1], 小畠秀和 [1], 後藤琢也 [1],
Kanta KAWAMOTO[1], Hidekazu KOBATAKE[1], Takuya GOTO[1]
[1] 同志社大学，Doshisha University#1,

1. Introduction

The heat of fusion of metals is a critical thermophysical property that governs the mass transfer phenomenon at the solid-liquid interface. Therefore, obtaining reliable data of heat of fusion is crucial for the numerical simulations of high temperature material processing via liquid phase such as the crystal growth or casting[1]). Conventionally, the heat of fusion is measured using thermal analysis such as differential scanning calorimetry with containers. However, the reactivity of metals with the container material can lead to sample contamination and increased uncertainty in the heat of fusion measurement. To address this challenge, the electrostatic levitation method has been employed for heat of fusion measurements by analyzing the temperature profile during recalescence[2]). However, on the ground condition, electrostatic levitation has a difficulty in the controlling the atmosphere because measurement is needed to be conducted under vacuum. In this study, we conducted the heat of fusion measurement for Fe, Ni, and Co using a closed type aerodynamic levitator with controlling the oxygen partial pressure.

2. Principle of measurement

When recalescence was occurred at the temperature of the hypercooling limit, the heat of fusion ΔH_f, can be expressed with the following equation,

$$\Delta H_\mathrm{f} = C_\mathrm{p}\Delta T_\mathrm{hyp}, \tag{1}$$

where, C_p denotes the heat capacity at constant pressure, ΔT_hyp is hypercooling limit.

On the other hand, when the solidification processed with no undercooling, the heat of fusion is expressed with the following equation, with considering both radiative heat loss and forced convection heat transfer,

$$\Delta H_\mathrm{f} = \int -\left\{\frac{\sigma_\mathrm{B} A \varepsilon_\mathrm{T}}{m}(T^4 - T_\mathrm{r}^4) + h_\mathrm{g}(T - T_\mathrm{r})\right\} dt. \tag{2}$$

Here, m is the mass of sample, σ_B is Stefan-Boltzmann constant, A is surface area of the sample, ε_T is total hemispherical emissivity, T is temperature of the sample, T_r is the surrounding temperature, h_g is heat transfer rate.

Alternatively, when the solidification processed with undercooling, the heat of fusion is expressed with the following equation,

$$\Delta H_{\mathrm{f}} = C_{\mathrm{p}}\Delta T + \int -\left\{\frac{\sigma_{\mathrm{B}} A \varepsilon_{\mathrm{T}}}{m}(T^4 - T_{\mathrm{r}}^4) + h_{\mathrm{g}}(T - T_{\mathrm{r}})\right\} dt. \quad (3)$$

3. Experiment

Fe, Ni, and Co samples with the 2.0 mm diameter were levitated within Ar-H_2 gas flow using a closed type aerodynamic levitator. The samples were subjected to heating by CO_2 laser irradiation. Once the samples were completely molten, the laser was turned off and samples were cooled, and then solidified. Temperature of the samples was measured by two-color pyrometer that was calibrated using the Wien's law at the respective melting point.

4. Results and discussion

An example of the temperature profile of Fe during solidification is shown in **Figure. 1**. From the temperature profile, the undercooling, ΔT, and the thermal plateau time, Δt, can be obtained. The correlation between ΔT and Δt of Fe sample is shown in **Figure. 2**. Since the term of $\frac{\sigma_{\mathrm{B}} A \varepsilon_{\mathrm{T}}}{m}(T^4 - T_{\mathrm{r}}^4) + h_{\mathrm{g}}(T - T_{\mathrm{r}})$ in Eq (2) or Eq (3) cab be considered as constant when the temperature was kept the melting point, the correlation between ΔT and Δt has a linear function as shown in **Figure. 2**. By the extrapolating the linear relation of ΔT vs. Δt liner function to $\Delta t = 0$ ΔT_{hyp} can be determined. From **Figure. 2**, the ΔT_{hyp} of Fe was determined as 280 ± 16 K. When the C_{p} of 45.4 J K^{-1} mol^{-1} in the literature value[3)] and the determined ΔT_{hyp} of 280 K were substitute in Eq (1), ΔH_{f} of Fe was determined to be 12.7 ± 0.7 k J mol^{-1}. By conducting similar measurement ΔT_{hyp} of Ni and Co were determined to be 414 ± 46 K and 360 ± 30 K, respectively. These ΔT_{hyp} values coupling with the literature C_{p} values[4)] gave ΔH_{f} of Ni and Co as 16.1 ± 1.8 k J mol^{-1}, and 14.6 ± 1.2 k J mol^{-1}, respectively. These ΔH_{f} values of Fe, Ni and Co measured using closed type aerodynamic levitator showed a good consistence with the literature values[4)] within the uncertainty.

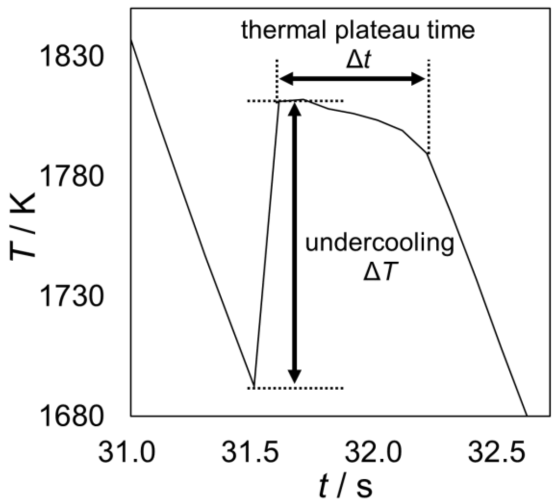

Figure 1. An example of temperature profile during solidification.

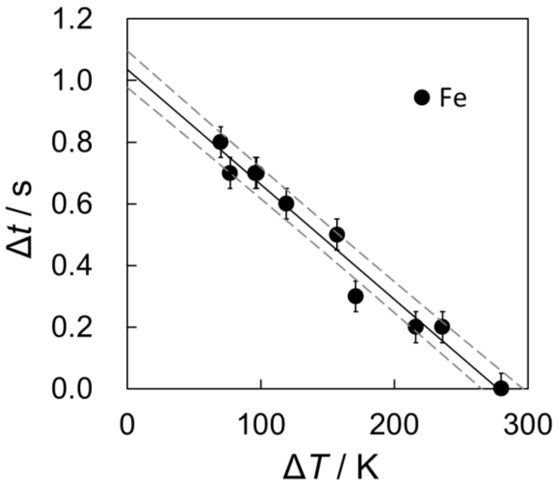

Figure 2. Correlation between undercooling and thermal plateau time of Fe.

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