The experimental design of the density of liquid Sn and liquid Ge from the transparency of γ rays

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Abstract

Density measurement by the high-energy γ ray attenuation method was briefly summarized. The results obtained for molten Ge and Sn up to 1200℃ were presented in comparing with those measured with other techniques.

§1. Introduction.

Several different techniques exist to measure the densities of liquid metals and alloys. Among them, Archimedean method, pycnometric method, maximum bubble method are quite often used. Although less familiar, the high-energy γ ray attenuation method has a number of advantages as compare with the above-mentioned techniques.

1. A sample sealed in a molten quartz ampoule can be used and hence the oxidation and/or preferential evaporation of constituent elements can be minimized.
2. Since the γ ray beam penetrates the bulk of the specimen, the effects caused by the surface tension are not involved.
3. A fairy small amount of material (~2cm^3) suffices to do experiments.
4. Measurements can be done automatically and therefore high quality data can be obtained with less elaboration.

On the other hand the accuracy is limited by the statistics of the γ ray counting. It is not easy to obtain the data with the relative accuracy within to 10^{-3} in a finite time interval. However it should be emphasize that this accuracy can be comparable to and even better than the one obtained with other methods.

The mass density ρ_{alloy} of an alloy may be evaluated from equation (1) by measuring the linear attenuation coefficient μ_{alloy}, (2).

$$\rho_{alloy}=\mu_{alloy}/\mu^*_{alloy}, \qquad (1)$$

and

$$\mu_{alloy}=d^{-1}\ln(N/N_0) \quad (2)$$

where d is the thickness of specimen (or sample cell), N and N_0 are the number of γ rays passing through a cell of thickness d filled with a specimen and an empty sample cell, respectively. The mass absorption coefficient μ^*_{alloy} may be calculated with equation (3) in terms of the mass absorption coefficient μ^*_i, atomic mass M_i and fraction x_i of a constituent element i

$$\mu^*_{alloy} = \Sigma\mu^*_i M_i x_i/\Sigma M_i x_i \quad (3)$$

It should be noted that the mass absorption coefficient μ^*_i is the intrinsic unique constant of an element that is independent of temperature, pressure as well as the chemical environment. The molar volume, V, may be evaluated from the mass density, ρ_{alloy} with equation (4).

$$V =\Sigma M_i x_i/\rho_{alloy} \quad (4)$$

§2. Experiments.

As mentioned in the introduction, the mass absorption coefficient is a unique constant for each element. A value compiled in a standard reference book may be used for experiments under ideal experimental condition; i.e. for a slab with infinite size (Storm and Israel 1970). In this experiment μ^*_i was determined with the present experimental set-up using a powder specimen compressed in a steel tube for Ge and a single crystal rod for Sn (Tsuchiya 1991, Kakinuma and Tsuchiya 1996). The numerical figures of $\mu^*_i M_i$ used are ,

Ge: 5.0494 ± 0.0006 [cm^2]
Sn: 8.7433 ± 0.0008 [cm^2]

A high purity Ge and Sn (5N grade) enclosed in a fused quartz ampoule were used. The shape of the quartz ampoule is schematically shown in the Figure 1.

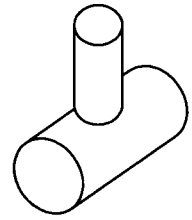

Fig. 1. Cylindrical quartz cell, walls are about 1.1 mm in thickness. Dimensions of absorber volume: length d is about 2.5 cm and diameter is about 1.1 cm

About 110 MBq of 137 Cs was used as a γ ray source of 662 keV. The experimental procedure was as follows.

1. The thickness, d, of an ampoule was determined by measuring the attenuation coefficient of mercury enclosed in it. The thickness was 2.5 ~ 2.7 cm. The dependence of d on the temperature was neglected because the thermal expansion of fused quartz is as small as 10^{-3} with respect to the one for molten Ge and Sn.
2. In the temperature interval between the melting point and a maximum temperature, the γ ray counting for 40 min. was repeated for several times at every 10 or 20 K, and then the whole procedure was cycled a few times.
3. The dependence on the temperature of the background attenuation of γ rays, referred to as f(T), caused by the furnace and the empty cell was measured independently and it could be fitted to a linear temperature dependence.

$$f(T)=1+0.7788693\times10^{-5}(T-T_0) \qquad (5)$$

4. The counting rate, $N(T)$, obtained in the step 3 was converted to the molar volume using the following relation.

$$V(T)=-C/\ln\{N(T)/N_0(T_0)f(T)\}, \qquad (6)$$

where $C=d\mu^*_{alloy}\Sigma M_i x_i$, and $N(T_0)$ is the counting rate for the background at $T=T_0$, respectively. Typically N_0 was ~ 1.5×10^6 for 40 min. and $N(T)$ was 4~ 6×10^5 for 40 min.

§3. Results

Figures 2 and 3, restively, show the results for Ge and Sn. The error caused by the statistics of the γ ray counting is given by $N(T)^{-1/2}$, and which can account for the scatter of the data. The absolute error estimated from the difference in the data obtained with a different sample was within to 0.5%. In the figures the data compiled in review papers are also shown

for comparison. The data so far reported for Sn are reasonably in agreement with the present results while the data for Ge show fairly large scatter. (Iida and Guthrie 1988, Crawley 1974)

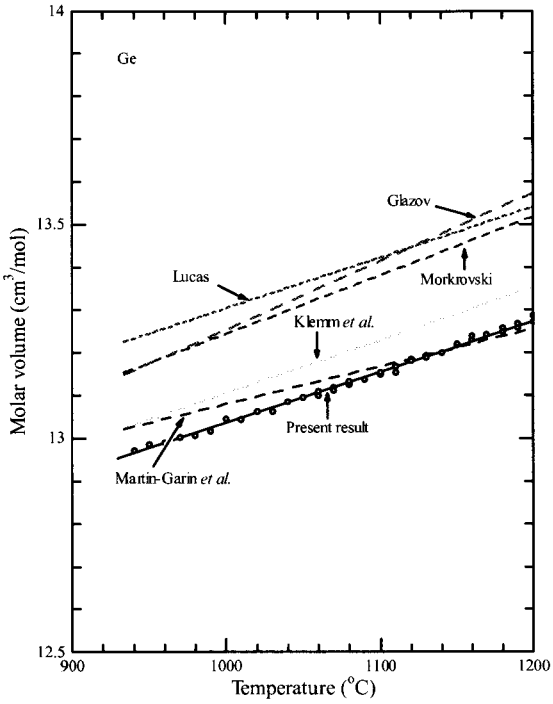

Fig. 2. Molar volume of Ge as a function of temperature.

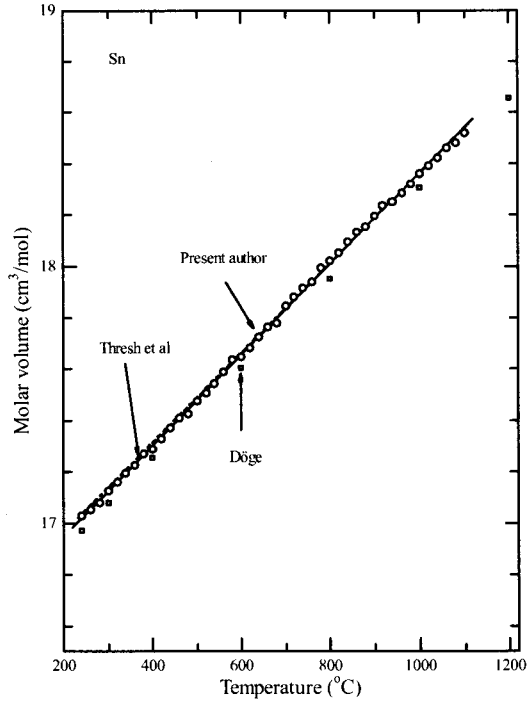

Fig. 3. Molar volume of Sn as a function of temperature.

§5. Experimental design of the density of liquid in much higher temperatures

In the atmospheric pressure, it was found that the fused quartz cell changed its shape slightly after measurements at temperatures higher than 1200℃. This deformation causes systematic error in the density measurements using the γ ray attenuation method. The deformation of the fused quartz cell can be avoided to some extent if the measurements are carried out *in vacuo* or in the low-pressure inert gas.

For measurements at high temperatures a specially designed furnace, which enables measurements *in vacuo*, was made with financial support from NASDA. The photograph shows the vacuum chamber with water jacket made by Wada Stainless Steal Manufacturing Co. Ltd. The experiments with this furnace are now in progress.

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