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Excess Viscosity of Binary Molten Alloys[†]

Toshio ENJYO*, Takamichi IIDA** and Masahito KANAI***

Viscosity of molten metals and alloys is an important factor in welding or metallurgical processes. For example, the rate of dissolution at the interface between solid and molten metals is mainly determined by the viscosity of molten metals. Therefore numerous attempts have been made to derive an equation relating the viscosity of binary molten alloys to its component and the viscosities of its components, but none can be regarded as satisfactory.

Generally, viscosities A—B binary molten alloys are expressed in the three cases of (a), (b) and (c) as shown in **Fig. 1**. The experimental results for Cu—Sn and Bi—Sn alloy systems⁹ are shown in **Fig. 2** and **3**, respectively. The curves (a) and (c), i. e., deviation from the straight line (b) are termed as excess viscosity (η^E). An excess viscosity is defined by

$$\eta^E = \eta - x_A \eta_A - x_B \eta_B \quad (1)$$

where, η is viscosity of alloys, x is composition and the subscripts A and B refer to the components. η^E is defined as $+\eta^E$ when η is above the straight line (b), η^E is $-\eta^E$ when under the straight line (b). An excess viscosity is used to represent deviation from linear dependence of viscosity with composition.

In the case of real liquids, particularly alloy

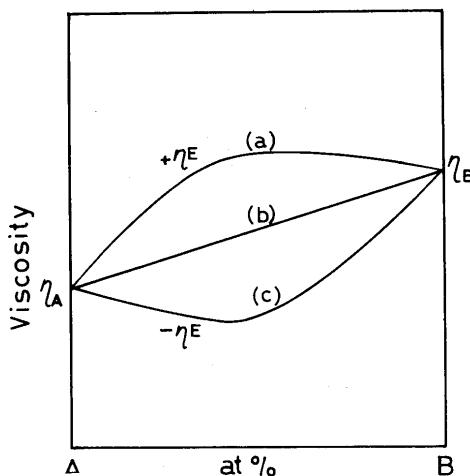


Fig. 1. Viscosities vs composition in the A—B binary systems.

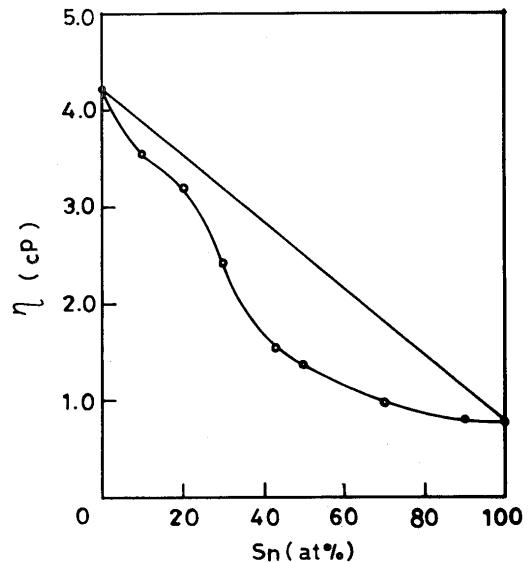


Fig. 2. Viscosities of molten Cu—Sn alloys at 1100°C. ($-\eta^E$)

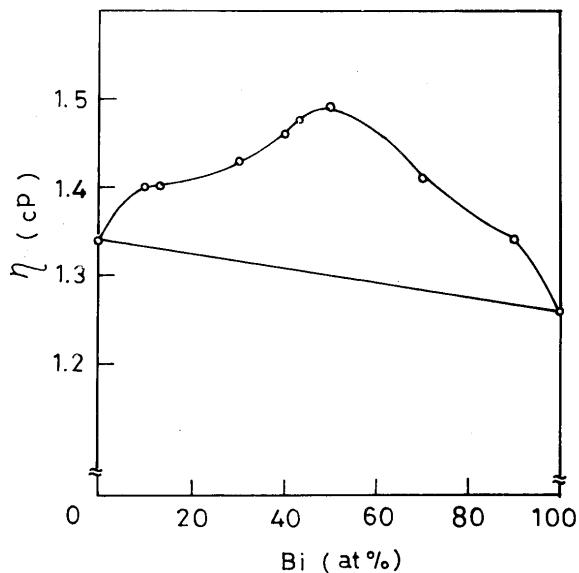


Fig. 3. Viscosities of molten Bi—Sn alloys at 400°C. ($+\eta^E$)

systems, it may be impossible to calculate theoretically because of complex interactions between atoms. But the structures of molten metals, i. e., radial distribution functions or intensity patterns, which are usually obtained by X-ray (neutron) diffraction, can be calculated

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using a simple treatment such as a hard sphere model.²⁾ Therefore it is expected that the structure sensitive properties such as viscosity and diffusion based on ionic motion may be determined approximately by aid of an adequate hard sphere model. The ionic motion is supposed to be impeded by frictional forces. The friction constant (γ) consists of two parts due to the soft part (γ_s) and the hard part (γ_h). They are expressed by the following simple relation

$$\gamma = \gamma_s + \gamma_h \quad (2)$$

Now, we neglect the soft part and take note of only the hard part. According to the reference 3),

$$\eta = \frac{1}{a} M^* \gamma_h = \frac{1}{a} M^* \frac{8}{3} \rho \sigma^2 g(\sigma) \sqrt{\frac{\pi kT}{M}} \quad (3)$$

where,

- a: average distance between the centers of atoms in molten state
- M^* : effective mass of an atom for diffusion
- ρ : number density ($=a^{-3}$)
- σ : hard sphere radius
- $g(\sigma)$: radial distribution function
- k: Boltzmann's constant
- T: absolute temperature (°K)
- M: mass of an atom

Supposing that the effective mass of an atom for diffusion is proportional to its real mass, the eq. (3) is rewritten by:

$$\eta \propto \frac{8}{3} \rho^{\frac{4}{3}} g(\sigma) \sqrt{\pi kT} \cdot \sigma^2 \sqrt{M} \quad (4)$$

Then, we assume that a hard sphere radius is equal to an ionic radius which has been presented by Pauling. Excess viscosities are expressed in terms of difference in ionic radii between the components.

The results are summarized in **Table. 1**. From the Table 1, it is evident that excess viscosities can be arranged by the difference in ionic radii. This results may be considered as follows. The effect of the hard sphere radius which may be a main term in eq. (4) is expressed by:

$$\begin{aligned} \eta^E &\propto x_A \sigma_A^2 + x_B \sigma_B^2 - (x_A \sigma_A + x_B \sigma_B)^2 \\ &= x_A x_B (\sigma_A - \sigma_B)^2 = x_A x_B (\Delta\sigma)^2 \end{aligned} \quad (5)$$

That is, η^E tends to become less negative as the difference between σ_A and σ_B , $|\sigma_A - \sigma_B|$, increases. On the contrary, concerning the effect of mass of an atom, η^E tends to become positive as the difference between M_A and M_B , $|M_A - M_B|$, increases. As can be seen from the Table 1, the excess viscosity of Pb-Bi alloy system is negative. For this reason, it is considered that, though the difference in ionic radii between Pb and Bi is small, the excess viscosity is

Table 1. Relationships between excess viscosities and difference in ionic radii between components.

Molten alloys	$\Delta\sigma (A)$	η^E
Au-Sn ¹⁾	0.66	—
○ Ag-Sb	0.64	—
○ Ag-Sn	0.55	—
Al-Cu ²⁾	0.46	—
Au-Cu ³⁾	0.41	+ -*
K-Na ⁴⁾	0.38	—
Cd-Sb ⁵⁾	0.35	—
○ Cu-Sb	0.34	—
Ag-Cu ¹⁾	0.30	—
○ Hg-In	0.29	—
○ Cu-Sn	0.25	—
Al-Zn ⁶⁾	0.24	—
○ Cd-Bi	0.23	—
K-Hg ⁷⁾	0.23	+
Pb-Sb ⁸⁾	0.22	—
Mg-Pb ⁹⁾	0.19	+
Na-Hg ¹⁰⁾	0.15	+
Al-Mg ²⁾	0.15	+
Pb-Sn ⁸⁾	0.13	+
○ Sb-Bi	0.12	+ -*
Ag-Au ¹¹⁾	0.11	+
Pb-Bi ¹²⁾	0.10	—
○ In-Bi	0.07	+
○ Sn-Bi	0.03	+

○: measured by S. Takeuchi and T. Iida

(*): Excess viscosities change from positive to negative with composition

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determined by the $(\Delta\sigma)^2$, since the atomic mass of Pb is nearly equal to the atomic mass of Bi.

As $(\Delta\sigma)$ becomes smaller, η^E is found to change

from negative to positive. According to this result, its critical value is considered as $(\Delta\sigma) \approx 0.23$ (Å).

Though we have neglected the soft part and considered only hard part, a distinctive feature for the viscosities of molten alloys, i. e., excess viscosities can be estimated.

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