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The Absolute Thermoelectric Power of Some Liquid Bismuth-Based Solutions

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Abstract—The absolute thermoelectric powers of liquid Bi containing Mg, Hg, As, Te, Se and S have been measured within the temperature range 400–600 °C. The thermopower of liquid Bi is increased by the addition of S, Se, Te or Hg, whereas it is decreased on the addition of As or Mg. The trend of the electro-negativity dependence of these thermopowers is roughly parallel to that of the resistivities. The results are compared with calculations based on the Faber-Ziman theory using three theoretical pair structure factors. Although the agreement is good in resistivities, except for As, there appears to be a serious discrepancy in the case of thermopower.

1. Introduction

In our previous paper⁽¹⁾ we reported results for the resistivities of a series of liquid Bi-based alloys and compared them with those predicted on the basis of the substitutional model due to Faber-Ziman⁽²⁾, in which the alloy is assumed to possess a single structure factor for the ionic configurations. This paper reports some measurements on the thermoelectric power of the similar solutions; Mg, Hg, As, Te, Se, and S are dissolved into liquid Bi. The thermopower is closely related to resistivity, and the Faber-Ziman theory which accounts for the resistance characteristics should also be applicable to the thermoelectric properties. Further, three theoretical structure factors⁽³⁾ are used in the calculation of the effect of ionic configuration in the alloys.

2. Experimental

The method⁽⁴⁾ of measurement and the purity⁽¹⁾ of the Bi, Mg, Hg

As, Te, and S used were as described in our previous works. The Se was 99.999% pure.

3. Results

The experimental results obtained for the absolute thermoelectric power Q of Bi - 2 and 5 at .% M solutions are illustrated in Fig. 1.

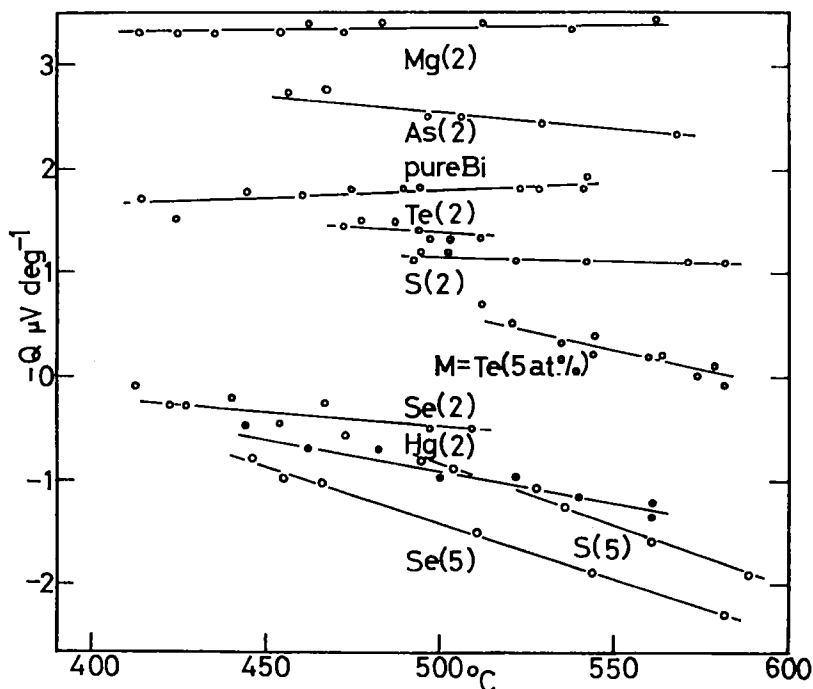


Figure 1. Absolute thermoelectric power Q $\mu\text{V deg}^{-1}$ of Bi-M solutions.

The temperature coefficients change from slightly positive value in pure Bi to negative, except in the case of Bi-Mg. The increase in Q due to the addition of another element can be seen in Bi-Hg, Te, Se and S solutions and Q changes from negative to positive. As can be seen in Fig. 2, the change in thermoelectric power is positive when the resistivity change is positive, and vice versa.

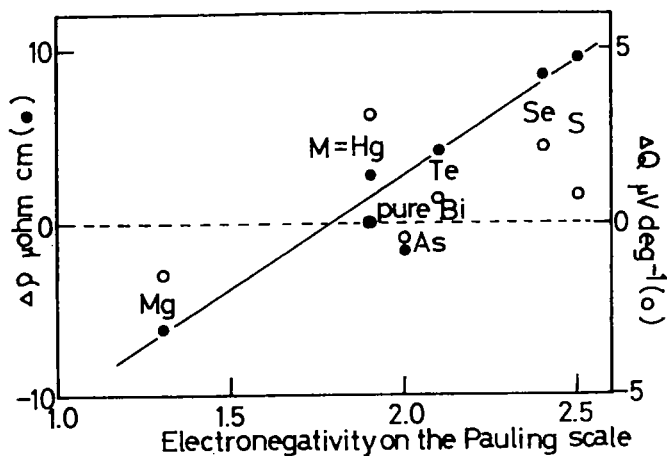


Figure 2. Relation between electronegativity and thermoelectric power change or resistivity one in Bi-2 at .% M solutions at 550°C, $\Delta Q = Q - Q_0$ (pure Bi) $\mu\text{V deg}^{-1}$ or $\Delta\rho = \rho - \rho_0$ (pure Bi) $\mu\text{ohm cm}$.

4. Discussion

These data may be compared with calculations based upon the Faber-Ziman theory,⁽²⁾ involving the three partial structure functions S_{00} , S_{11} and S_{01} for 0-0, 1-1 and 0-1 pairs, in which 0 and 1 denote the solvent and solute species, respectively. We used the Percus-Yevick theory to calculate S_{00} , S_{11} and S_{01} under the approximation of the rigid sphere model⁽³⁾ for binary liquids with the total packing fraction η adjusted, as well as possible, to fit the observed X-ray data of pure liquid Bi and corrected to allow for changes in volume due to the solute. If we restrict the calculation to dilute solutions of the order of 2 at .% solute or less, it can be proposed that interactions between solute atoms, and between solvent atom and solute one are negligible ($S_{11} \approx 1$ and $S_{01} < 0.2$, respectively), and that no structure of liquid metal is significantly altered by the addition of solutes up to 2 at .% ($S_{00} \approx S_0$ (pure metal)). The scattering due to individual ions can be interpreted in terms of the point charge model⁽⁵⁾ for anions and Animalu's pseudopotentials⁽⁶⁾ for cations, but adjusted for changes of volume and Fermi energy.

Theoretical and experimental values for the thermoelectric power change ΔQ ($\equiv Q - Q_0$ (pure metal)) and the resistivity change

$\Delta\rho$ ($\equiv \rho - \rho_0$ (pure metal)) are compared in Table 1, where the experimental results of resistivities obtained by the same authors⁽¹⁾ are also included. In $\Delta\rho$ there is no significant disagreement between theory and experiment, except for As. By contrast, the solute dependence of ΔQ is different from that expected on theoretical grounds. For this discrepancy two possible explanations would be

TABLE 1 Comparison between Theoretical and Experimental Values for ΔQ $\mu\text{V deg}^{-1}$ and $\Delta\rho$ $\mu\text{ohm cm.}$, Produced by 2 at. % of Impurity in Liquid Bi at 550 °C

Solute	Z†	Theory			Exptl.	
		ΔQ	$\Delta\rho$	$\alpha\dagger$	ΔQ	$\Delta\rho$
Mg	2	-3.1	-6.1	0.95	-1.5	-6.2
Hg	2	-0.2	2.7	0.895	3.1	2.8
As	5	-0.2	3.9	1	-0.5	-1.7
Te	6	-0.1	3.2	0.86	0.9	4.1
Se	-2	-0.4	9.0	1	2.2	8.5
S	-2	-0.2	9.9	1	0.8	9.4

† Z is the valency for the positive ions. The -2 state is assumed for the charge of the negative ions in the group VI elements.

‡ α is the hard-sphere ratio: $\sigma_\alpha/\sigma_\beta$ ($0 \leq \alpha \leq 1$) with diameter σ_α .

(a) the inadequacy of the Faber-Ziman theory and (b) energy-dependent pseudopotentials. In view of this basically successful theory we prefer to focus attention on (b). However, no quantitative calculations⁽⁴⁾ can now be carried out from an adequate analytical form of the energy dependent Heine-Animalu potentials⁽⁷⁾ because of the lack of accurate values of parameters of these elements.

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