Viscosity of Bi-Ga liquid alloys

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Abstract. The viscosity of Bi$_{1-x}$Ga$_x$ liquid alloys has been studied by means of oscillating crucible method. Temperature dependences of viscosity reveal the Arrhenius-like behavior at elevated temperatures and deviate from it at ones close to miscibility gap curve. The results on viscosity are analyzed from the point of view of concentration fluctuations.

1. Introduction
The binary Bi-Ga system reveals the miscibility gap in liquid state in the significant concentration range (from 38.5 to 91.5 at.% Ga). The critical point, according to recent publication, has coordinates: 535K and 70 at.% Ga [1]. Recently, the interest of researchers has been focused on studying the concentration fluctuations near the critical point. These studies were carried out by different methods: x-ray and neutron diffraction [2, 3], viscosity, electrical resistivity, ultrasonic measurements as well as thermodynamic ones [4-9]. The results indicate anomalous behavior just above critical point showing the existence of large scale concentration fluctuations.

Nevertheless the interpretation of experimental data is only qualitative in many respects. The relation between structural changes and physical properties is limited and needs more quantitave characteristics not only for the alloys of this system but generally for all binary melts with miscibility gap.

Analyzing the data on viscosity for this system, one can see that in many works the major attention is drawn to the concentration dependence. Temperature dependence is analyzed less, especially above phase separation line, where transition from two-phase liquid to monophase one occurs.

Besides, the published experimental data reveal some disagreement because in liquids of such kind the measurements are very sensitive to thermal prehistory. On that reason in this paper we present the results on viscosity measurements of Bi$_{1-x}$Ga$_x$ molten alloys.

2. Experimental procedure
The ingots of Bi-Ga binary system were prepared from pure Bi and Ga of 99.99% purity. The viscosity coefficient was measured with an oscillating–cup viscometer. The samples were placed into the chamber filled with helium of high purity; they were homogenized for 5 hour at the highest temperature. The viscosity was measured during cooling the sample, values of viscosity coefficients were calculated from measured logarithmic decrements and the period of oscillations by Roscoe’s equation [10]. The accuracy was less than 3%.
3. Results and Discussion
The temperature dependences of viscosity for Bi$_{1-x}$Ga$_x$ liquid alloys are shown in figure 1. As it seen the viscosity decreases gradually according to exponential law. In case of alloys corresponding to miscibility gap (x=0.10; 0.30; 0.50) the anomalous behavior reveals within some definite temperature regions. One can see that at temperature decreasing the viscosity begins deviate from exponential law showing the excess values. Such viscosity increase reaches a maximum value at some temperature and then decreases. The main sharp maximum is observed for the melt of critical composition. The temperature dependence of viscosity for alloy of this composition was compared with ones obtained in [4] (figure 2).

**Figure 1.** Viscosities of liquid Bi, Ga and five melts of the Bi-Ga system.

**Figure 2.** Comparison of our values of viscosity with obtained in [3] for liquid Bi$_{30}$Ga$_{70}$.

One can see that coincidence between two curves occurs within wide temperature range, except the region of anomalous behavior. May be this discrepancy is caused by different rates of cooling.

The temperature dependences of viscosity for the alloys containing 61.5 and 80 at.% Bi have no such pronounced maxima on the background of exponential functions as for previous melts. But some features are revealed in these functions also. We suggest that deviation from exponential dependence at temperatures about 550K is caused in this case also by concentration fluctuations which have significantly less scale, but promote formation of Ga-Ga and Bi-Bi microgroups.

**Figure 3.** Natural logarithm of viscosity for liquid Bi$_{10}$Ga$_{90}$, Bi$_{30}$Ga$_{70}$, Bi$_{50}$Ga$_{50}$ (a) and Bi$_{61.5}$Ga$_{38.5}$, Bi$_{60}$Ga$_{20}$ (b) alloys.

For more detail analysis of viscosity behaviour in the regions were anomalies are pronounced we plotted the temperature dependences of viscosity in logarithmic scale

\[
\ln \eta = f \left( \frac{10^3}{T} \right)
\]
The curves, obtained in this way (figure 3), allowed us to determine the temperature points where the experimental data begin deviate from the linear dependence. These temperatures were determined for the melts with concentration within miscibility gap as well as for ones, whose concentration is out of it.

The dependence of this characteristic temperature ($T_{ch}$) vs Ga content (figure 4) shows in fact the same values within the miscibility gap, and significantly less for alloys containing 20 and 90 at.% Ga. These data allowed us to state that large concentration fluctuations occur at temperatures higher than miscibility gap curve or liquidus line. Nevertheless it should be noted that in the melts, which concentration is out of the miscibility gap, these fluctuations are less than in the alloys corresponding to the range where two demixing liquids coexist.

![Figure 4](image1.png)  
**Figure 4.** Concentration dependence of $T_{ch}$ and phase diagram.

![Figure 5](image2.png)  
**Figure 5.** Concentration dependences of viscosity for $\text{Bi}_{1-x}\text{Ga}_x$ liquid alloys.

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**4. Conclusions**

The temperature dependences of viscosity in $\text{Bi}_{1-x}\text{Ga}_x$ liquid alloys show the Arrhenius like behavior at elevated temperatures and anomalous behavior revealed as local maximum at temperatures close to phase separation point. This maximum decreases gradually with heating.

Such maximum has the main value in the melt of critical composition and this is caused by significant concentration fluctuations. Small scale concentration fluctuations exist also in molten alloys, which compositions are outside of the miscibility gap.

**References**

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