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Influence of Ni-atoms addition on the structure of In-Ga-Sn liquid ternary eutectic alloy

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Abstract. The influence of Ni-atoms on the structure of In-Ga-Sn eutectic melts has been studied by means of X-ray diffraction method. The structure factors and pair correlation functions were calculated. The analysis of main structure parameters, obtained from these functions showed that Ni-atoms are diluted in structural units of liquid eutectic melt.

1. INTRODUCTION

Liquid eutectic alloys with low temperature melting point are widely used as solder materials, heat transfers, matrix in composite systems. In recent years the liquid In-Ga-Sn alloy of ternary eutectic concentration attracts the attention of researches as component of liquid magnetic systems, especially of ferro-colloid ones [1]. Besides, this liquid eutectic alloy is promising as a reference temperature point with corresponding definite melting temperature. Taking into account these facts it is interesting to study the influence of other elements on the structure and properties of In-Ga-Sn liquid eutectic alloy. First of all there is of great interest to study the addition of elements with ferromagnetic properties influence on structure of this eutectic melt. It has a practical motivation due to that some part of ferromagnetic colloidal particles suspended in $\text{In}_{20.497}\text{Ga}_{66.960}\text{Sn}_{12.543}$ liquid eutectic matrix upon some time period of composite system existence undergoes the diluting process.

This work represents the results on influence of Ni-atoms on the structure of In-Ga-Sn ternary melt at temperatures higher than melting temperature.

2. EXPERIMENTAL

Both samples — pure eutectic melt and added with 5 at% of Ni were prepared by melting in furnace filled with pure argon. There were used the elements of high purity for synthesis of samples (99.999% - Ga, In, Sn, 99.9% - Ni).

X-ray diffraction studies were carried out with using of high temperature diffractometer. Specimen was placed in vacuum chamber filled with He of high purity in order to avoid its oxidation. X-ray were monochromatized by LiF single crystal installed in initial beam. Scattering intensities were recorded with step 0.05° and counting time of impulses was equal 100s in each angular point. Temperature was measured with accuracy $\pm 2\text{K}$.

Angular dependencies of scattered intensity were corrected on absorption, polarisation, incoherent intensity and anomalous dispersion and then normalized to electron units. Corrected intensity curves were used to calculate the structure factors (SF) which were analyzed. Pair correlation functions (CF) were calculated from SF by means of Fourier transformation. Besides radial distribution functions of atomic density $G(r)$ were estimated too. From these graphical functions the main structure parameters were estimated as positions of principal peaks in SF's — S_1 , S_2 , second to first positions ratio (S_2/S_1), height of main peak $\{a(s_1)\}$, main peak positions r_1 and r_2 and squares under principal peaks in CF's. Last of them determine the most probable interatomic distances and number of neighbour atoms.

For calorimetric studies [2] the differential scanning calorimeter Pyris-1 DSC (Perkin-Elmer) was used. The heating rate was equal to 1 K/min.

3. RESULTS AND DISCUSSION

SF of liquid In-Ga-Sn eutectic alloy at temperature 5K above melting point is shown in Fig.1. As it can be seen its profile has a feature in a principal peak. Its right hand side reveals a shoulder, what is a characteristic of SF for liquid tin. This shoulder is commonly interpreted as an evidence of covalent bonding between some atoms in liquid state what is the reason of inhomogeneous atomic distribution. Talking into account our results on the structure of this melt obtained early [3] in our works we consider the structure of liquid eutectic alloy as consisting the self-associated atoms of indium and randomly distributed atoms of Ga and Sn. The (Ga+Sn) atomic solution is more responsible for total atomic arrangement in melt than the self-associated In-In atomic groups. This assumption follows from analysis of main structure parameters (table 1) which are similar to values of corresponding ones of liquid Ga and Sn.

Upon addition of Ni atoms the most significant changes are pronounced in principal peak profile (Fig.1, table 1). Particularly its height decreases and s_1 parameter shifts towards corresponding parameter of liquid Ni. The shoulder on right hand side also exists in structure factor of Ni-doped melt. This shoulder should be more pronounced if suppose that Ni-atoms are self-associated, but it is no observed in profile of first peak. Therefore, it is possible to suggest that Ni-atoms are diluted in self-associated structural units of other component of eutectic melt. This suggestion is in agreement with equilibrium phase diagrams Ni-Ga, Ni-In and Ni-Sn, where the tendency to preferred interaction of unlike kind atoms is pronounced in chemical compounds existence [4]. The solubility of Ni in In, Ga and Sn is very small but these semimetals show the higher solubility in Ni, especially far Ni-Ga system.

On the other hand the analysis of pair correlation functions allowed us to conclude about the better agreement of experimental value of r_1 to corresponding one of self-associated

atomic distribution model than for random distribution one (table 1) for both melts – initial and Ni-doped.

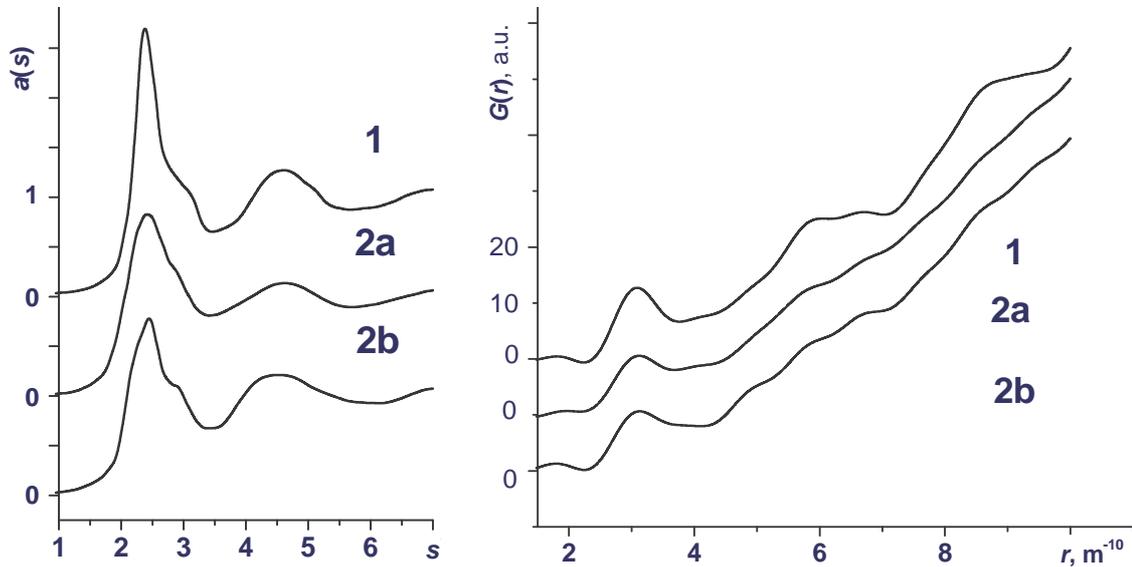


Figure 1. Experimental SF $a(s)$ and radial distribution functions $G(r)$ for: {1} In-Ga-Sn eutectic alloy at the 290K and In-Ga-Sn + 5 at% Ni studied alloys. {2a} – at the 870K and {2b} — at the 770K.

Model values of r_1 for random distribution $\{r_1^{R.D.}\}$ and self-associated $\{r_1^{S.A.}\}$ models were calculated according to formulas:

$$r_1^{S.A.} = \sum_i c_i k_i^2 r_i; \quad r_1^{R.D.} = \left(\sum_i c_i k_i \right)^2 \sum_i c_i r_i$$

where

- c_i – atomic fractions of components;
- i – number of components;
- k_i — scattering densities;
- r_i – most probable distances to first neighbours of components

Temperature increase of Ni-doped melt to 870K does not lead to significant changes of structure. Main peaks in structure factors and pair correlation functions decrease their height and become of more width. Such behavior is the evidence of topological disordering of atomic distribution. The transformation of chemically ordered distribution of atoms to random one is observed.

In order to check that molten alloy does not deviate from eutectic composition we have investigated it by means of high temperature calorimeter method (Figure2). As it can be seen there is a maximum without shoulder what confirms that concentration of this alloy corresponds to eutectic point.

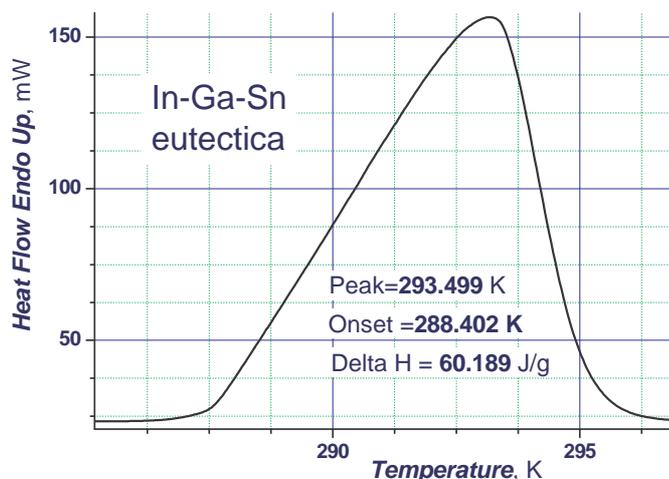


Figure 2. Results of calorimetric studies of In-Ga-Sn liquid eutectic alloy.

4. CONCLUSIONS

The addition of Ni-atoms to In-Ga-In liquid eutectic alloy promotes the formation of chemically ordered atomic distribution in structural units without significant changes in total topology of atomic arrangement. Temperature increase leads to topological disordering and formation of random atomic distribution.

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